Supporting Information

Syntheses, structural variation and characterization of a series of crystalline coordination compounds with 4-benzene-1,2,4-triazole: Polymorph, Incomplete spin transition and Single Crystal to Single Crystal Transformation

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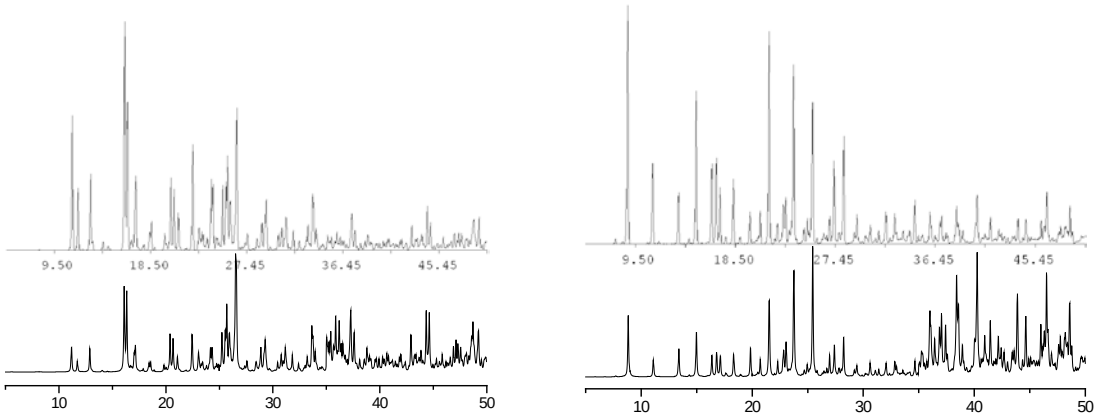
Table S1. Selected hydrogen bond lengths [Å] and angles [°] for **1-7**.

Donor-H···Acceptor	D-H	H···A	D···A	D-H···A
1				
C(7)-H(7)···Br(2)	0.95	2.81	3.640(4)	146
C(10)-H(10)···Br(2)	0.95	2.92	3.843(4)	165
2				
C(16)-H(16)···Br(1)	0.95	2.68	3.617(7)	169
C(15)-H(15) ···Br(2)	0.95	2.92	3.860(7)	171
3				
N2-H2B···O1	0.8600	2.2517	3.1088	174
C8-H8A···S1[a]	0.9300	2.8222	3.6587	150
C15-H15A···O2[b]	0.9300	2.3675	3.2722	164
4				
C1-H1-S1[a]	0.95	2.86	3.7302	153
C8-H8-O1	0.95	2.20	3.0891	162
C13-H13···N2[b]	0.95	2.52	3.4442	161
C14-H14···O1	0.95	2.36	3.2741	165
C16-H16···S4[c]	0.95	2.72	3.6689	175
C22-H22···S3	0.95	2.61	3.5179	161
C23-H23···S2[d]	0.95	2.81	3.7179	159
C28-H28···S3	0.95	2.75	3.6529	159
O1-H1A···S4	0.86	2.61	3.4548	169
5				
C1-H1-S2[a]	0.93	2.86	3.7345	168

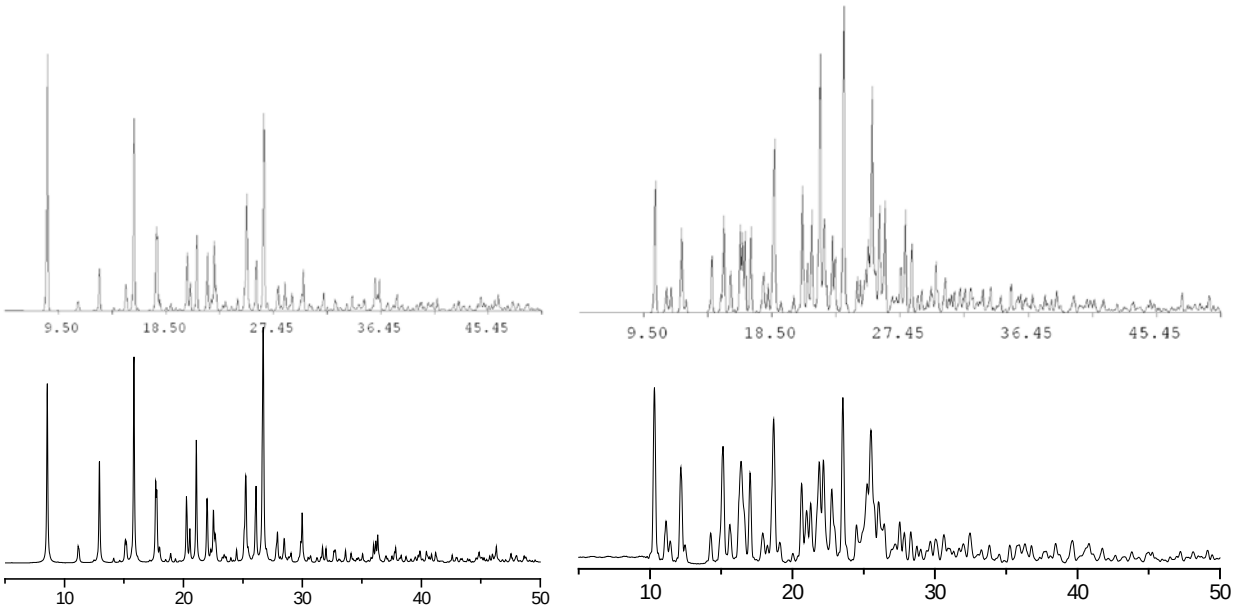
C9-H9-S4[b]	0.93	2.61	3.5136	164
C25-H25···S3	0.93	2.69	3.6125	173
C33-H33···O2[c]	0.93	2.23	3.1336	165
C34-H34···O1	0.93	2.35	3.2698	172
C36-H36···O1	0.93	2.47	3.4209	173
C39-H39···N1	0.93	2.54	3.4307	161
O1-H1A···S2[d]	0.82	2.43	3.3087	167
6				
O1-H1A···O2[a]	0.84	1.94	2.7668	171
O2-H2···N1	0.84	2.11	2.8281	143
C4-H4···O4	0.95	2.25	3.403	167
C22-H22···O4	0.95	2.53	3.3562	145
C15-H15···S4[b]	0.95	2.71	3.6517	172
7				
O(1)-H(1A)···O(6)[a]	0.85	1.95	2.7655	160
O(2)-H(2A) ···O(5)[b]	0.85	2.08	2.7306	132
O(2)-H(2B) ···O(25)	0.85	1.82	2.6665	179
O(19)-H(19A)···N(12)	0.85	2.09	2.9064	161
O(21)-H(21B)···O(10)[c]	0.85	2.04	2.7699	144
O(26)-H(26A) ··O(12)	0.85	1.99	2.8382	172
O(26)-H(26B)···N(29)	0.85	2.12	2.9279	159

Symmetry codes: For **3**: *a*: 1/2-x, 1/2-y, 1-z; *b*: 1/2-x, -1/2+y, 1/2-z. For **4**: *a*: 1/2+x,1/2-y,1-z; *b*: 1/2-x,1-y,1/2+z; *c*: -1/2+x,3/2-y,1-z; d: x, y, z+1; *d*: -x,1/2+y,1/2-z. For **5**: *a*: -1/2+x,1/2-y,2-z; *b*: 2-x,-1/2+y,3/2-z; *c*: 1/2+x,3/2-y,2-z; d: x, y, z+1; *d*: x,y,-1+z. For **6**: *a*: x,1+y,z; *b*: 1-x,1-y,1-z. For **7**: *a*: 1-x,1-y,-z; *b*: 1-x,1-y,1-z; *c*: x,-1+y,z.



(a) (b)

Fig. S1 X-Ray powder patterns (a) for **1** and (b) for **2**: (up) calculated on the basis of the structure determined by single-crystal X-ray diffraction. (down) X-ray diffractograms of an original sample.



(a) (b)

Fig. S2 X-Ray powder patterns (a) for **3** and (b) for **4**: (up) calculated on the basis of the structure determined by single-crystal X-ray diffraction. (down) X-ray diffractograms of an original sample.

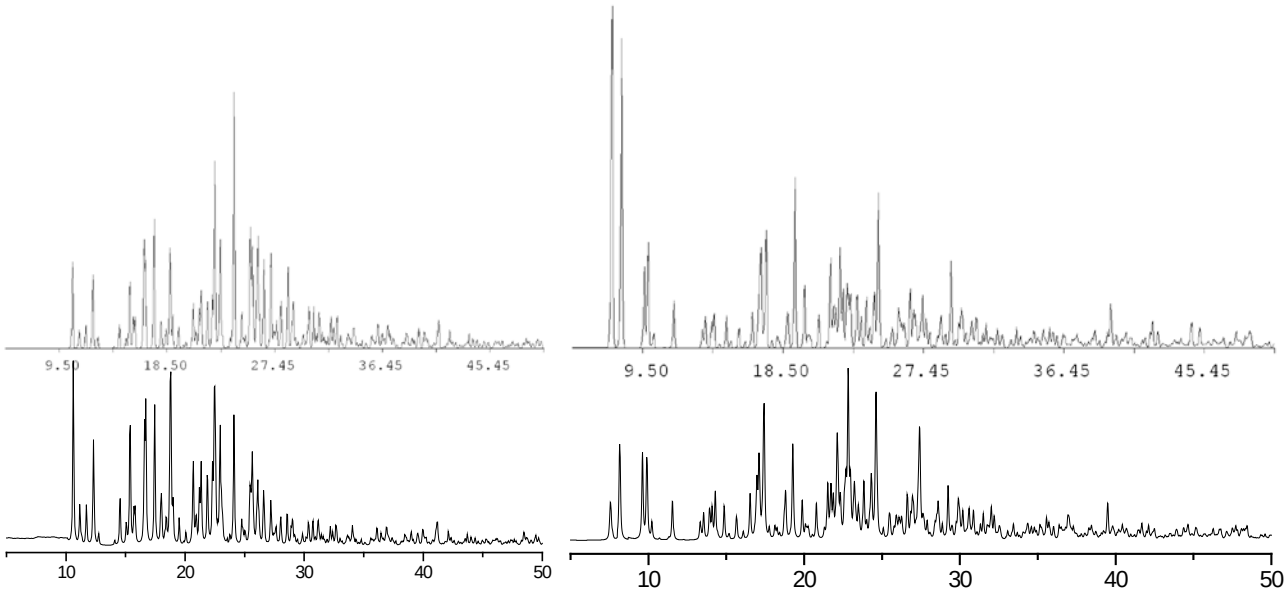


Fig. S3 X-Ray powder patterns (a) for **5** and (b) for **6**: (up) calculated on the basis of the structure determined by single-crystal X-ray diffraction. (down) X-ray diffractograms of an original sample.

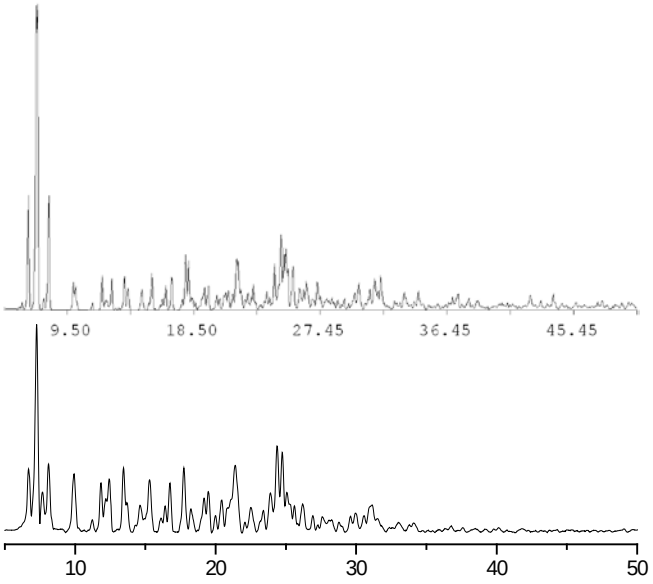
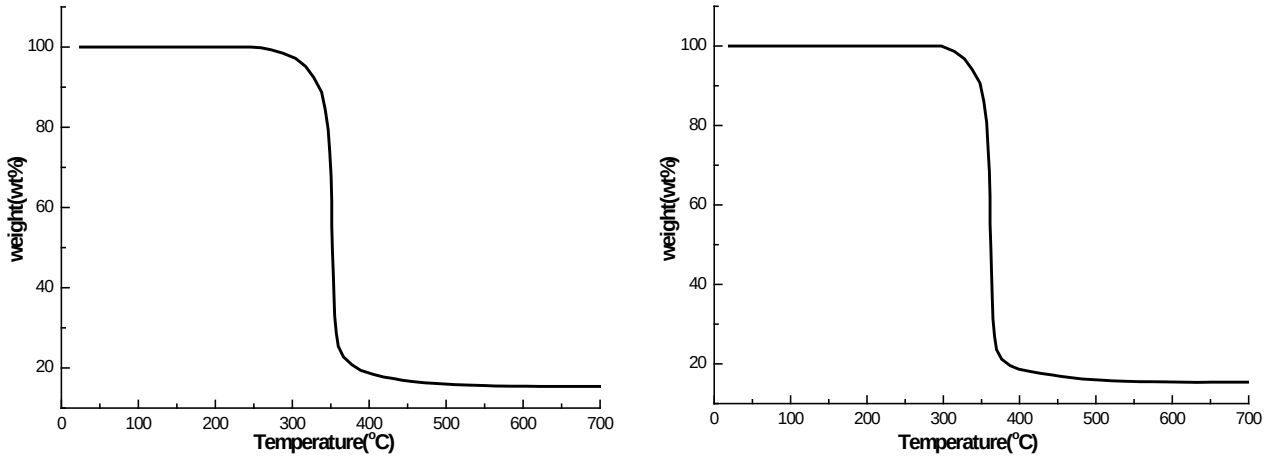
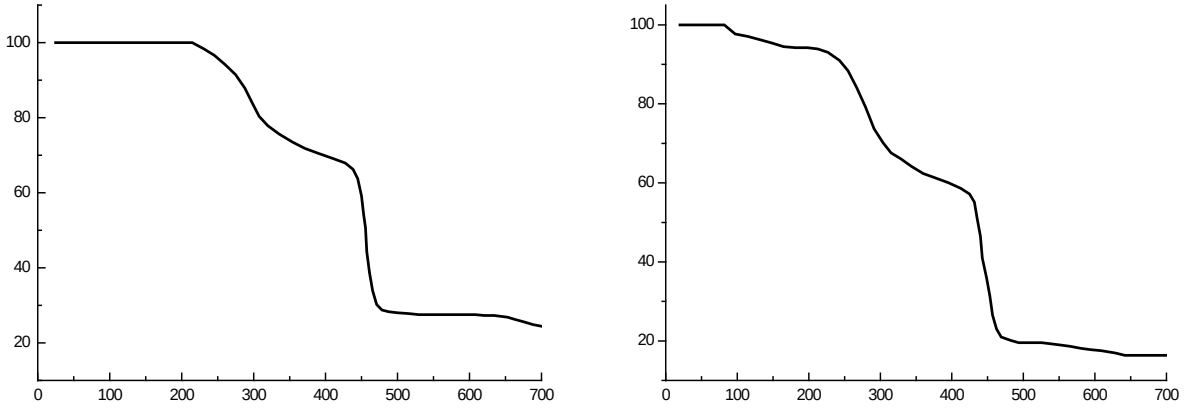


Fig. S4 X-Ray powder patterns for **7**: (up) calculated on the basis of the structure determined by single-crystal X-ray diffraction. (down) X-ray diffractograms of an original sample.



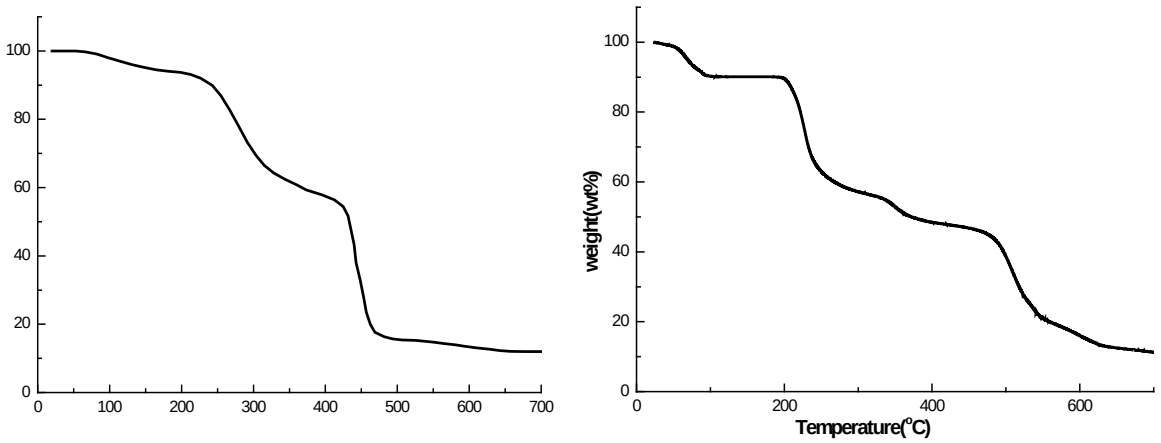
(a) (b)

Fig. S5 TGA curve (a) for **1** and (b) for **2**:



(a) (b)

Fig. S6 TGA curve (a) for **3** and (b) for **4**:



(a) (b)

Fig. S7 TGA curve (a) for **5** and (b) for **6**:

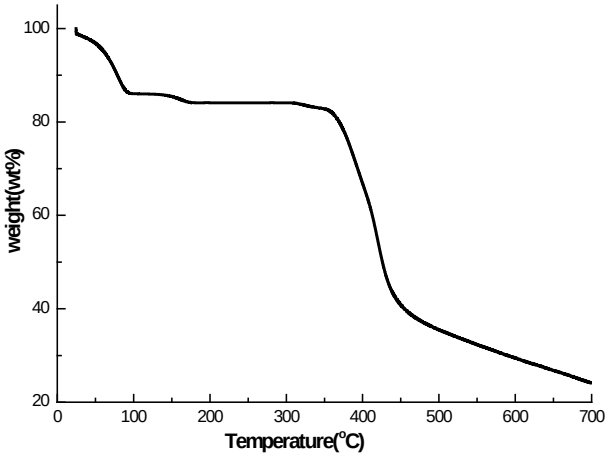


Fig. S8 TGA curve for **7**:

Some additional information for the fitting results for introducing the local anisotropy of the iron(II) ion in the calculation of XMT for complex **6**:

The fitting results for introducing the local anisotropy of the iron(II) ion in the calculation of XMT for complex **6:** We also attempted to introduce the local anisotropy of the iron(II) ion in the calculation of χ_M^D. This anisotropy splits the excited states in the zero field. We did not get any improvement of the fitting, which suggests that zero-field splitting of the excited states is small with regard to *J*.