Supplemental Section

S1: Single crystal analysis

Table S1: Selected bond lengths and angles for 1

Bond lengths /Å	
C(8)-Fe(1)	2.039(2)
C(9)-Fe(1)	2.041(2)
C(10)-Fe(1)	2.042(2)
C(11)-Fe(1)	2.039(2)
C(12)-Fe(1)	2.039(2)
C(13)-Fe(1)	2.018(3)
C(14)-Fe(1)	2.033(3)
C(15)-Fe(1)	2.050(3)
C(16)-Fe(1)	2.038(3)
C(17)-Fe(1)	2.033(3)
Angles / °	
N6-C7-C8	110.7(2)
C7-N6-N4	110.6(2)
C7-C8-C9	123.9(2)
C17-Fe1-C10	106.50(11)
Fe1-C8-C7	130.03(13)
Hydrogen Bonding /Å	
N6(H)…N2	3.101(2)
N2…C5(H)	3.378(7)
Ligand torsion/°	
N4-N6-C7-C8	178.7

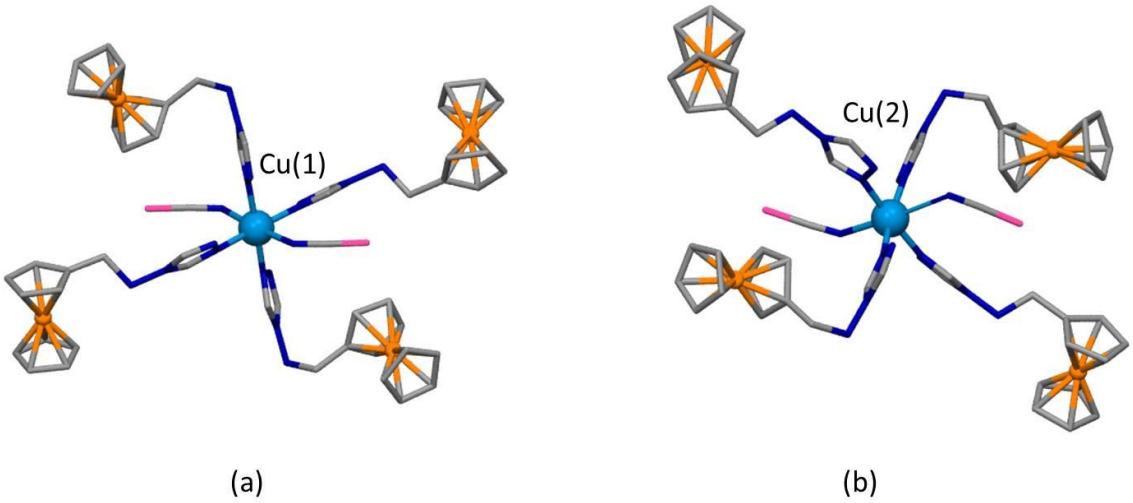

Figure S1: Comparison of the crystallographically distinct 1D chains in **5** containing either Cu(1) or Cu(2).

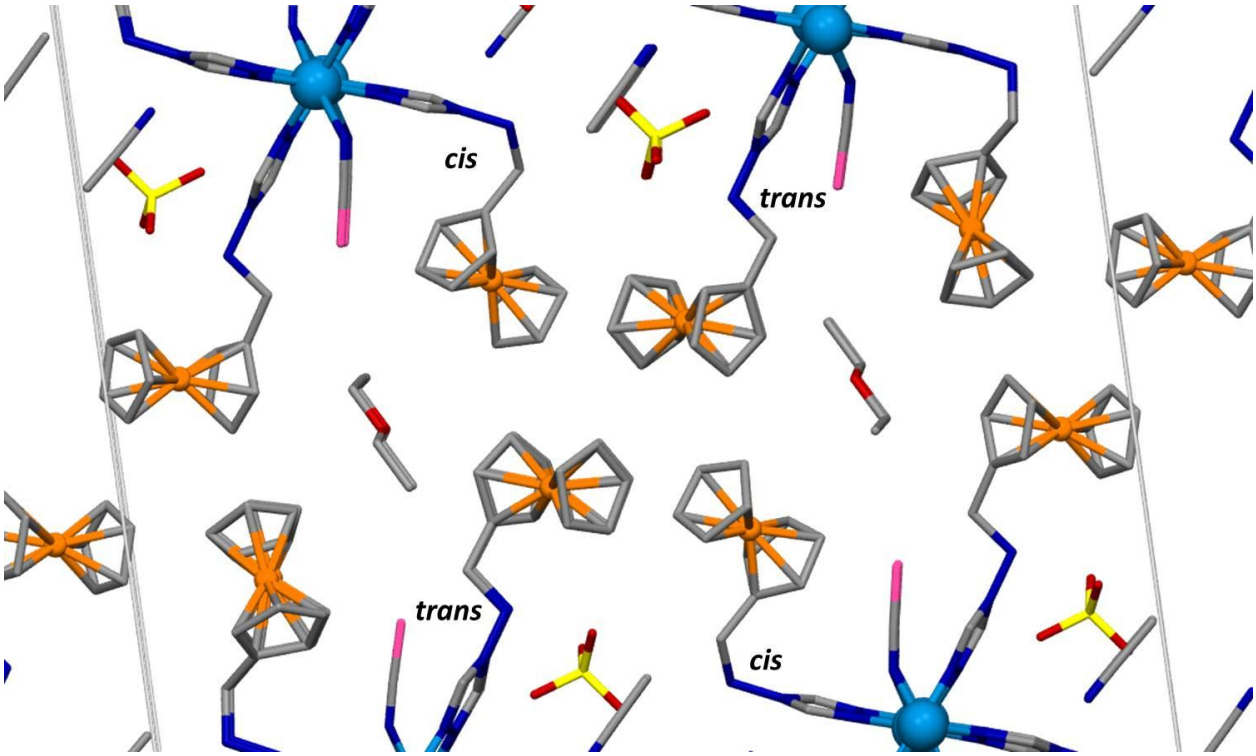

Figure S2: Image showing the *cis*- and *trans*-ATF ligands in **5** and their arrangement within the unit cell such that there are ferrocene rich areas between the chains. Viewed along the *b*-axis.

S2: Magnetic Susceptibility

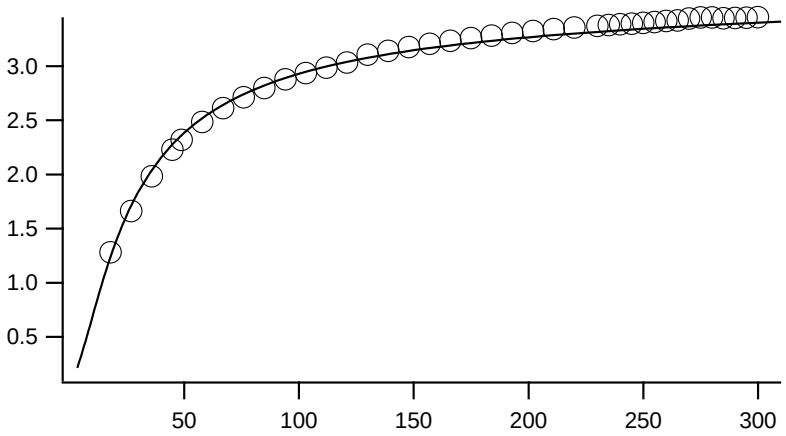

Figure S3: Plot of $\chi_M T$ versus temperature (K) for $[Fe(ATF)_3]\cdot(Br)_2\cdot 0.5(H_2O)$, **2**. The solid line is the best fit to a 1D Heisenberg $S = 2$ model (see text for parameter values).

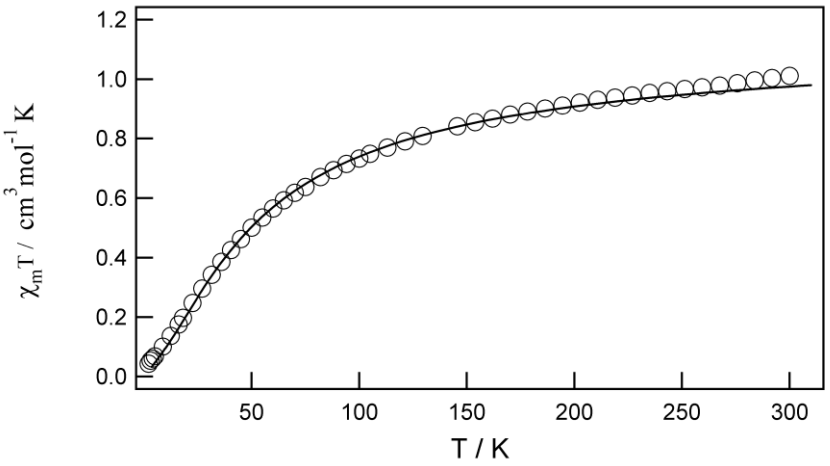

Figure S4: Plot of $\chi_M T$ versus temperature for $[Ni(ATF)_3]\cdot(ClO_4)_2\cdot 0.5(H_2O)$, **4**. The solid line is the best fit to a 1D Heisenberg $S = 1$ model (see text).

S5: Cyclic voltammetry

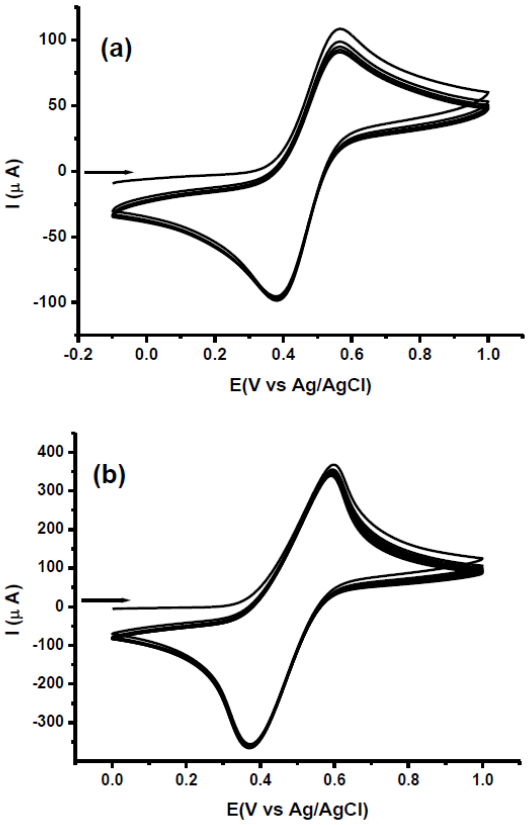

Figure S5. Multicyclic voltammograms obtained at scan rate of 100 mV s^{-1} for solid $[Fe(ATF)_3](ClO_4)_2 \cdot 0.5(H_2O)$, (a) and $[Ni(ATF)_3](ClO_4)_2 \cdot 0.5(H_2O)$, (b) mechanically attached to 1.5 mm diameter GC electrode when placed i