Supporting Information

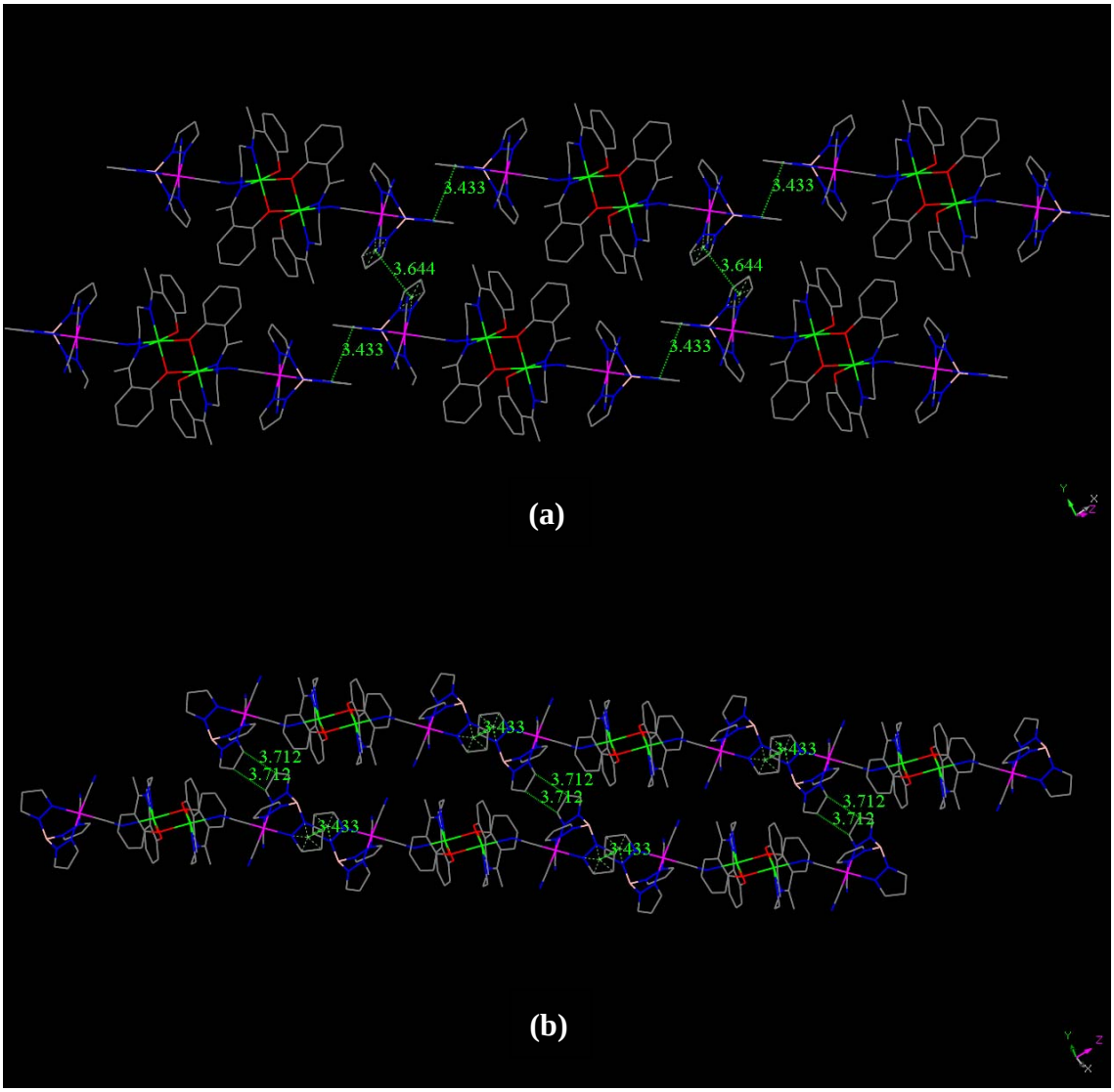


Fig. S1 Extended 3D structures of **1** constructed from π-π interactions between pyrazole rings of Tp ligands. (a) The noncovalent bonds with centroid distances of 3.433 and 3.644 Å lead to the formation of a 2D sheet. The shortest Fe-Fe distances are 7.752 and 7.775 Å through the respective intermolecular contacts. (b) The 2D architecture is formed by π-π stackings of the 1D chain with a centroid length of 3.433 and additional π-π forces between two aromatic carbon atoms of pyrazole rings with a separation of 3.712 Å through which the shortest Fe-Fe distance is 7.948 Å.

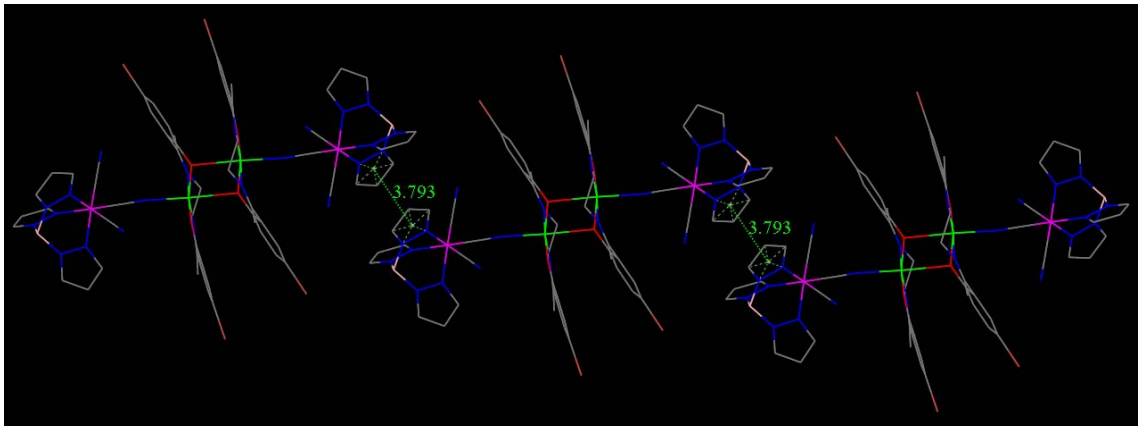
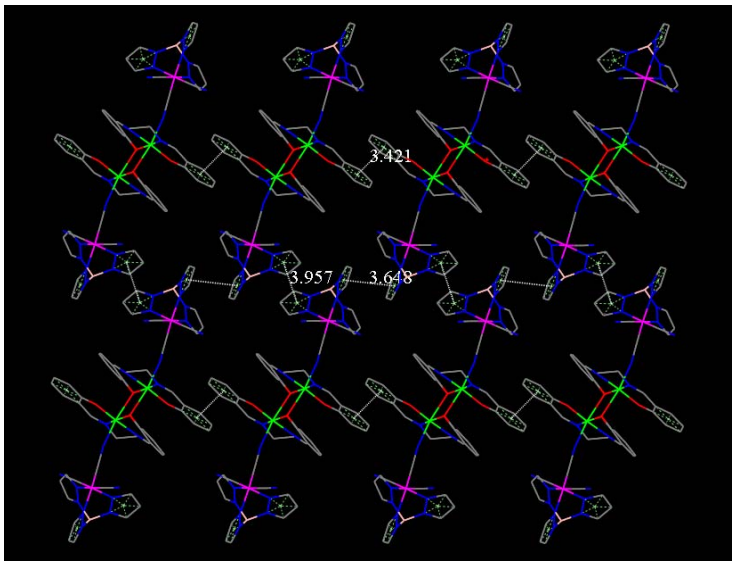
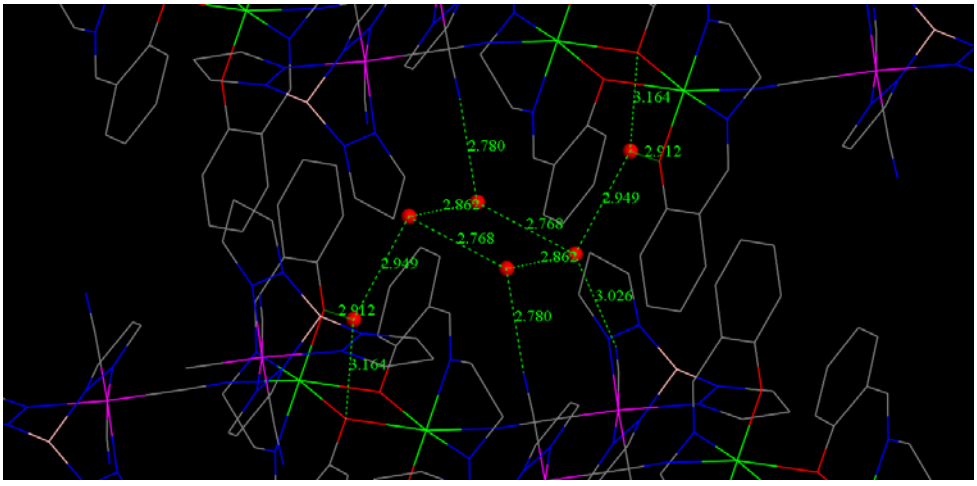


Fig. S2 Extended 1D structure of **2** showing π-π interactions between pyrazole rings of Tp ligands (centroid distance = 3.793 Å). The shortest Fe-Fe distance is 7.684 Å through the intermolecular contact.



(a)



(b)

Fig. S3 Extended structures of **3**. (a) The 2D layer is constructed by π-π interactions (dotted lines) between benzene groups of Schiff base ligands (centroid distance = 3.421 Å) and between pyrazole rings of Tp ligands (centroid distances = 3.648 Å and 3.957 Å). The shortest intermetallic distances are 9.204 Å through the benzene contacts and 7.713 Å via the pyrazole contacts. (b) The 2D sheets are interconnected via hydrogen bonds (dotted lines) among unbound CN groups, phenoxide oxygens, and lattice water oxygens, leading to the formation of a 3D network. The shortest M-M distance through the hydrogen bonds between the 2D sheets is 9.578 Å.

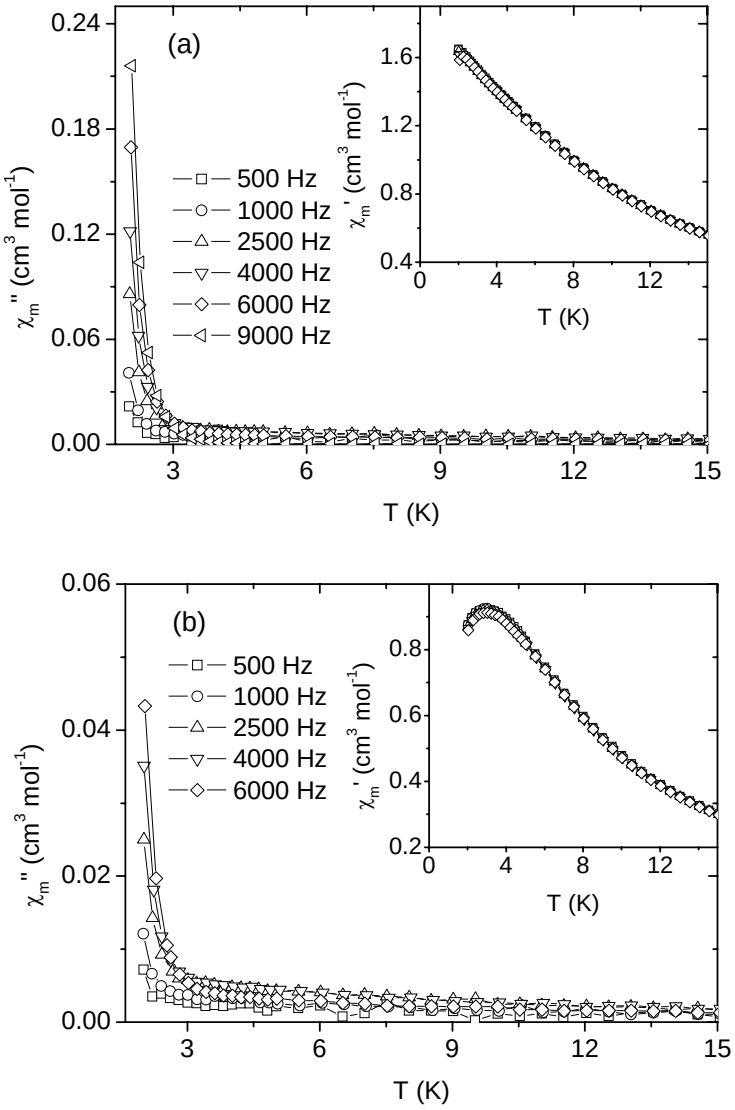


Fig. S4 Plots of in-phase (χ_m" in main panel) and out-of-phase (χ_m' in inset) components of ac magnetic susceptibility data for (a) **1** and (b) **2** at indicated frequencies.

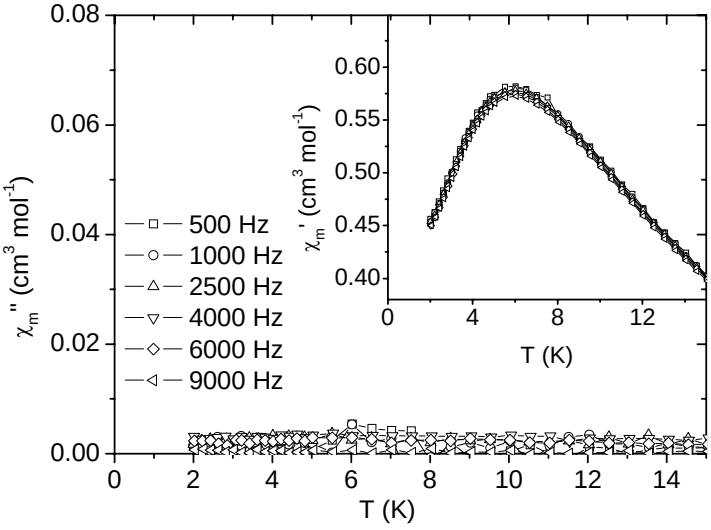


Fig. S5 Plots of in-phase (χ_m" in main panel) and out-of-phase (χ_m' in inset) components of ac magnetic susceptibility data for **3** at indicated frequencies.