Supporting information:

Bimetallic Cyanido-Bridged Magnetic Materials Derived from Manganese(III) Schiff-Base Complexes and Pentacyanidonitrosylferrate(II) precursor

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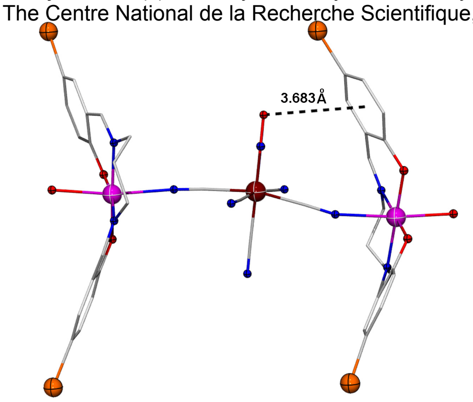


Figure S1: Ball-and-stick view of the environment of NO ligand in the trinuclear unit of complex **1**. The broken line suggests the possible intramolecular interactions between O_{nitro} and the aromatic ring of the Schiff base ligands.

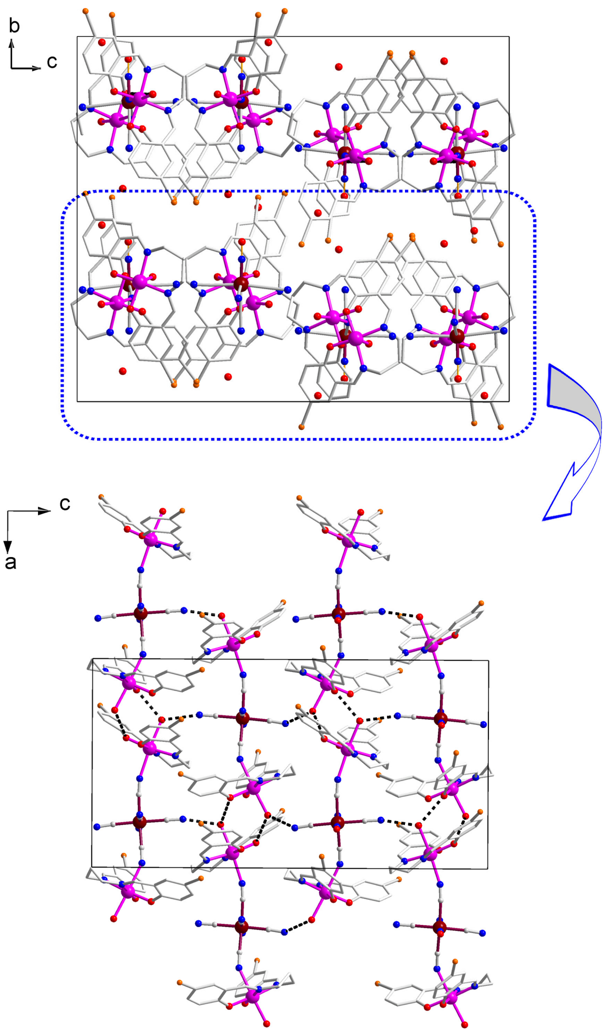


Figure S2: Packing arrangement of the 3-D supramolecular assembly of complex **1**. There is no obvious H-bonding interaction between two adjacent supramolecular layers.

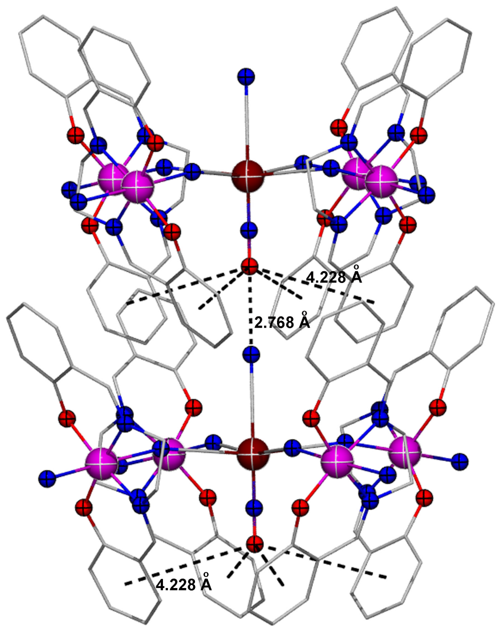
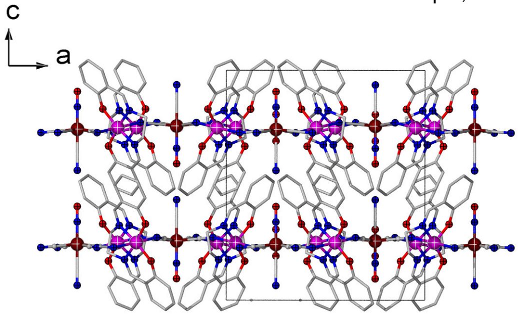
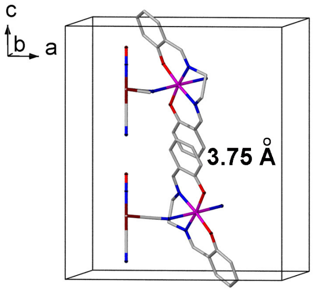


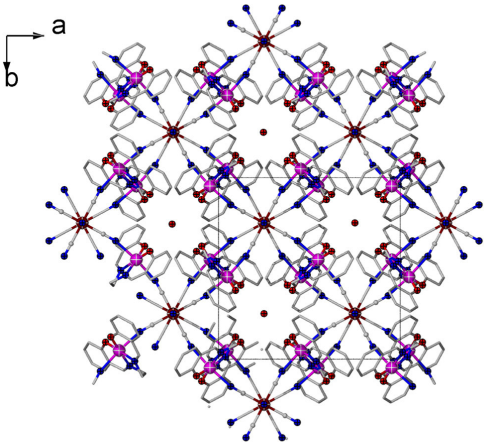
Figure S3: Ball-and-stick view of the environment of NO ligand in complex **2**. The broken lines suggest the possible intramolecular interactions between O_{nitro} and the aromatic ring of the Schiff base ligands, as well as the intermolecular interactions between O atom of the nitro ligand and adjacent N atom of the cyanido ligand.



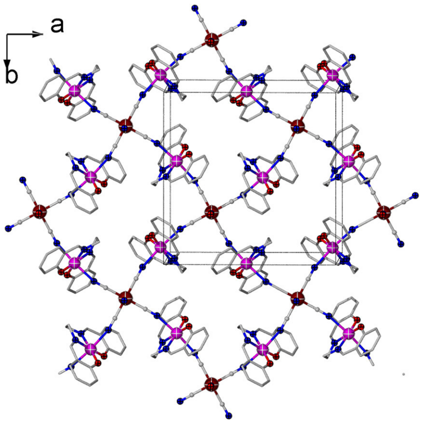
(a)



(b)



(c)



(d)

Figure S4: a) Packing arrangement of complex **2** viewed along *b* axis. (b) View of the π-π interaction between two aromatic planes derived from adjacent layers with the distance of ca. 3.75 Å. (c) Packing arrangement of complex **2** viewed along *c* axis. (d) View of the single 2-D layer in complex **2**.

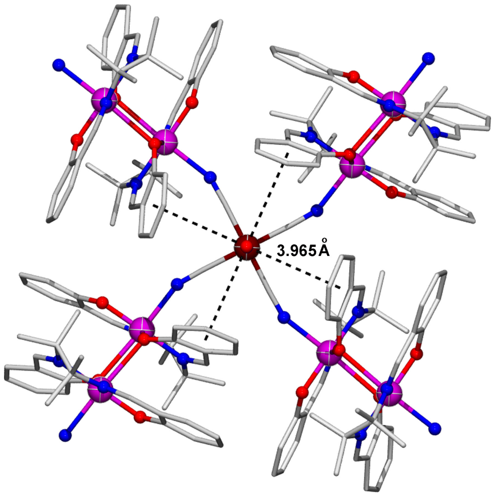


Figure S5: Ball-and-stick view of the environment of NO ligand in complex **3**. The broken lines suggest the possible intramolecular interactions between O_{nitro} and the aromatic ring of the Schiff base ligands.

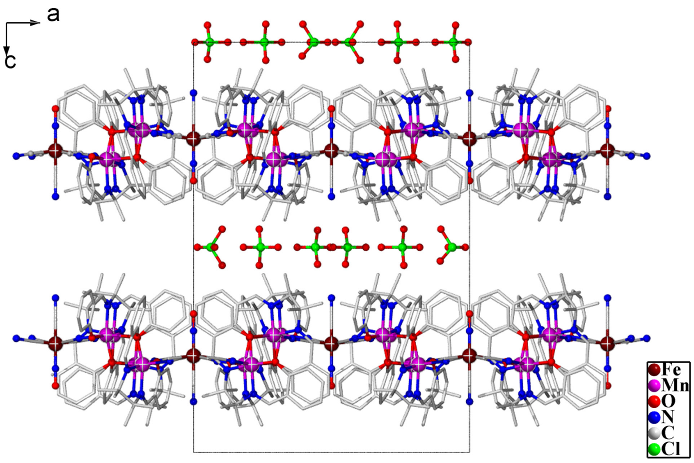


Figure S6: Packing arrangement of complex **3** viewed along *b* axis. H atoms and solvent water molecules are omitted for clarity. There are no direct H-bonding interactions between two adjacent layers.

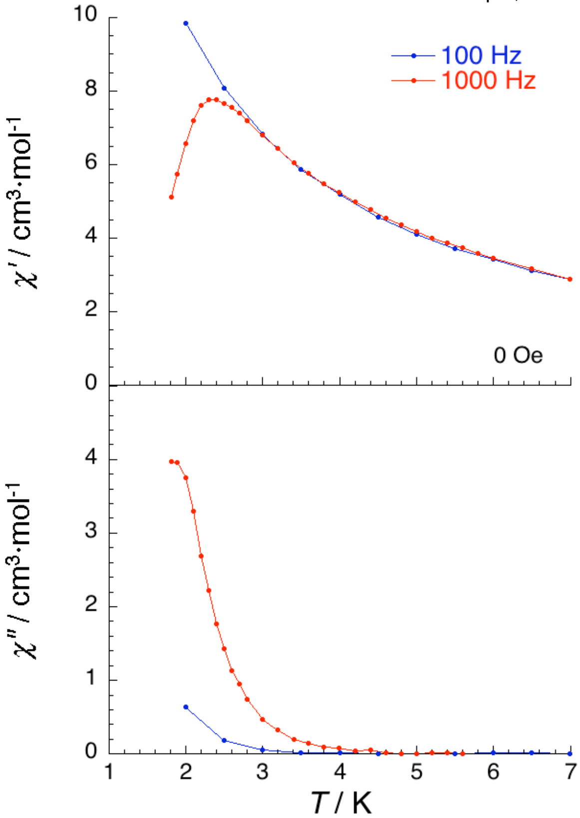


Figure S7: Temperature dependence at 100 and 1000 Hz of the in-phase (χ') and out-of-phase (χ'') component of the ac susceptibility for **3** below 7 K under zero dc-field.

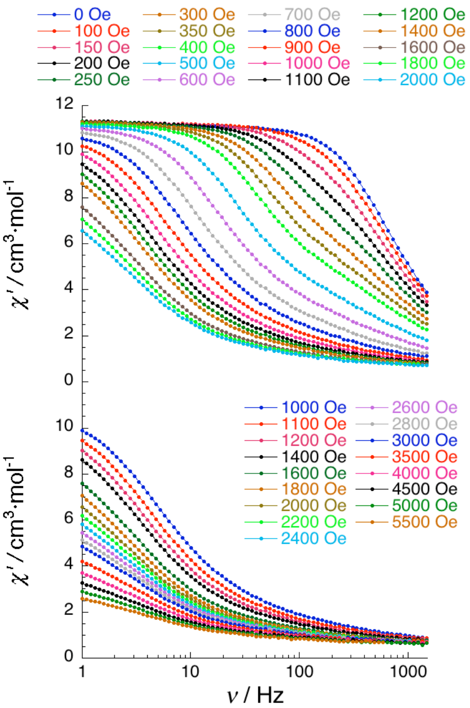


Figure S8: Frequency dependence of the in-phase component of the ac susceptibility for **3** as a function of the ac frequency between 1 and 1500 Hz at 1.8 K under dc-fields.

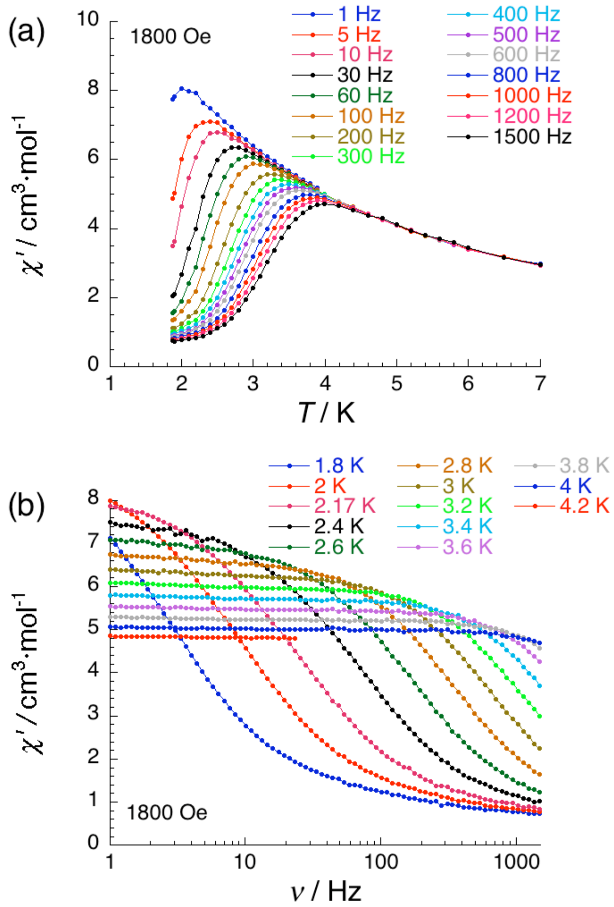


Figure S9: Temperature dependence at different ac frequencies (a) and frequency dependence at different temperatures (b) of the in-phase component of the ac susceptibility for **3** below 7 K under 1800 Oe.

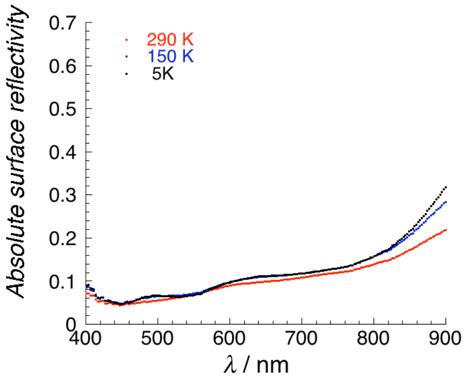


Figure S10: Absolute surface reflectivity *vs* wavelength for **1** at 290 (red), 150 (blue) and 5 K (black) using a white light of 0.5 mW/cm^2.

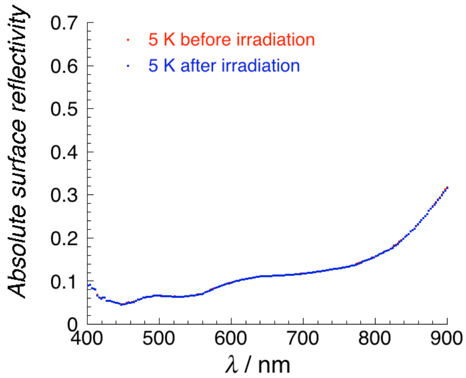


Figure S13: Absolute surface reflectivity *vs* wavelength for **1** at 5 K under a white light of 0.5 mW/cm^2: the red and blue dots are respectively before and after irradiation at 470 nm (60 mW/cm^2).

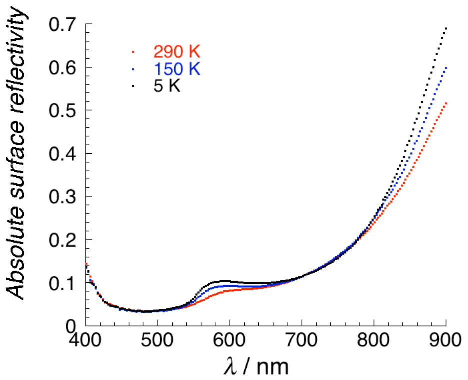


Figure S11: Absolute surface reflectivity *vs* wavelength for **2** at 290 (red), 150 (blue) and 5 K (black) using a white light of 0.5 mW/cm^2.

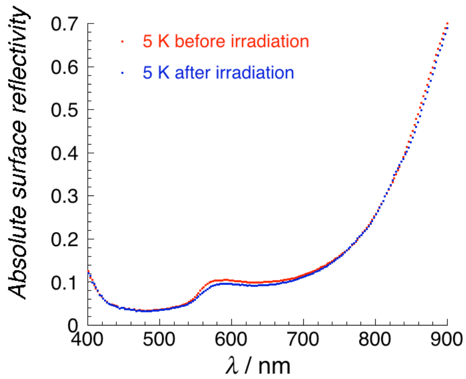


Figure S14: Absolute surface reflectivity *vs* wavelength for **2** at 5 K under a white light of 0.5 mW/cm^2: the red and blue dots are respectively before and after irradiation at 470 nm (60 mW/cm^2).

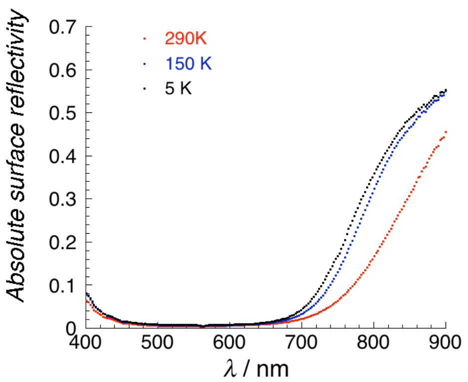


Figure S12: Absolute surface reflectivity *vs* wavelength for **3** at 290 (red), 150 (blue) and 5 K (black) using a white light of 0.5 mW/cm^2.

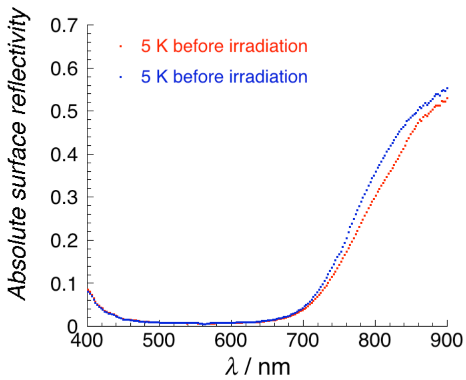


Figure S15: Absolute surface reflectivity *vs* wavelength for **3** at 5 K under a white light of 0.5 mW/cm^2: the red and blue dots are respectively before and after irradiation at 470 nm (60 mW/cm^2).