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\textsubscript{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\textsubscript{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.
**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 O₅ 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 ($\chi'$) and out-of-phase ($\chi''$) 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/cm².

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/cm².

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/cm².

Figure S13: Absolute surface reflectivity vs wavelength for 1 at 5 K under a white light of 0.5 mW/cm²; the red and blue dots are respectively before and after irradiation at 470 nm (60 mW/cm²).

Figure S14: Absolute surface reflectivity vs wavelength for 2 at 5 K under a white light of 0.5 mW/cm²; the red and blue dots are respectively before and after irradiation at 470 nm (60 mW/cm²).

Figure S15: Absolute surface reflectivity vs wavelength for 3 at 5 K under a white light of 0.5 mW/cm²; the red and blue dots are respectively before and after irradiation at 470 nm (60 mW/cm²).