Supplementary information

Highly Interface Dependent Spin Transport In Fe-Mn(DBTAA)-Fe Single Molecule Spintronic Device

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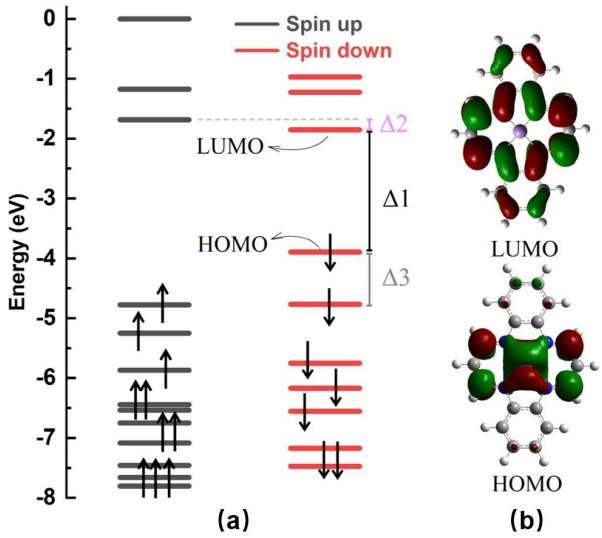

Fig. S1. (a) Schematic energy level diagram for the Mn (DBTAA) molecule. (b) The spatial distribution of LUMO and HOMO states of the isolated Mn(DBTAA) molecule. All simulations are based on Gaussian 16.

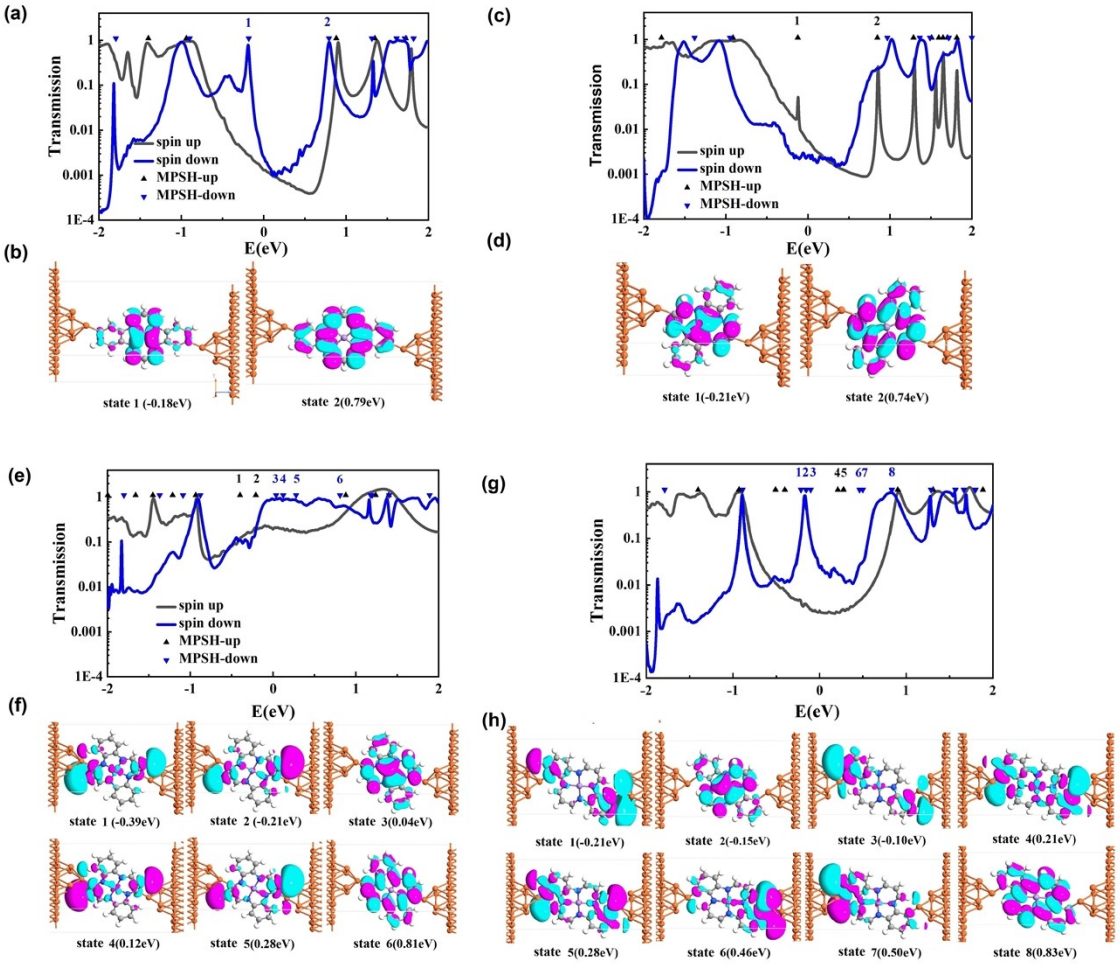

Fig. S2. Transmission spectra and corresponding distribution of transmission eigenstates for (a)-(b) C1, (c)-(d) C2, (e)-(f) C3 and (g)-(h) C4 configurations around the Fermi level. Therein, (b), (d), (f), and (h) show the transmission eigenstates corresponding to selected MPSH orbitals marked by triangles in transmission spectra of (a), (c), (e) and (g), respectively. The Fermi levels all are set to zero.

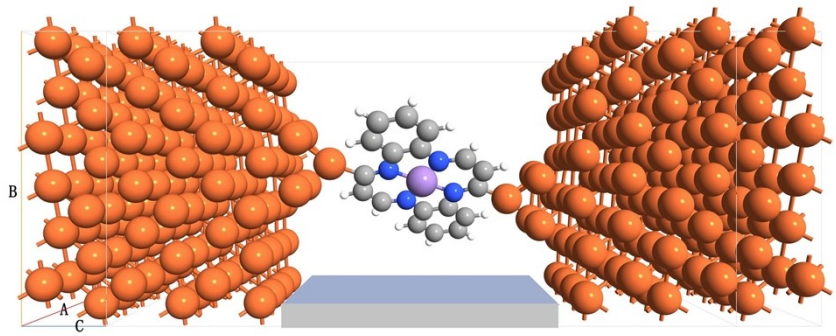

Fig. S3. The device structure of C3 configuration with an external gate electrode under the Mn(DBTAA) molecule. Note that the electrode Au superlattice is changed from triclinic to orthorhombic.

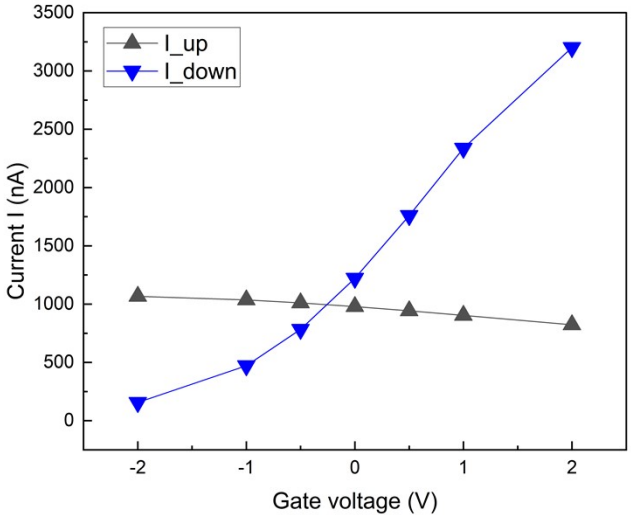

Fig. S4. Variation of spin-dependent current with gate voltage in C3 configuration. Here, the black regular triangles, blue inverted triangles represent the spin up and spin down current, respectively. The applied bias voltage is fixed as 0.1 V.

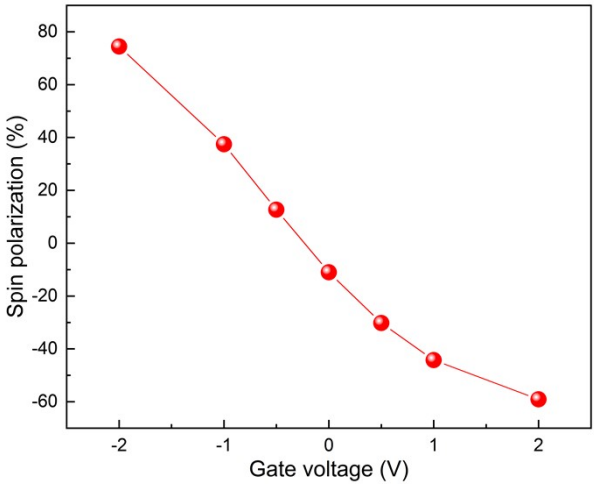

Fig. S5. Variation of current spin polarization with gate voltage in C3 configuration. The applied bias voltage is fixed as 0.1 V. Note that the solution of Poisson equation with gate electrode is different from that without gate electrode due to different boundary conditions used. For the former, the Neumann Boundary Condition is applied along the B lattice direction, while it is Periodic Boundary Condition in the latter. At the same time, the zero energy point of Fermi level is different in the two cases. Therefore, the calculated SP at zero gate voltage (-11%) is distinct from the case without placing a gate electrode (-60%).

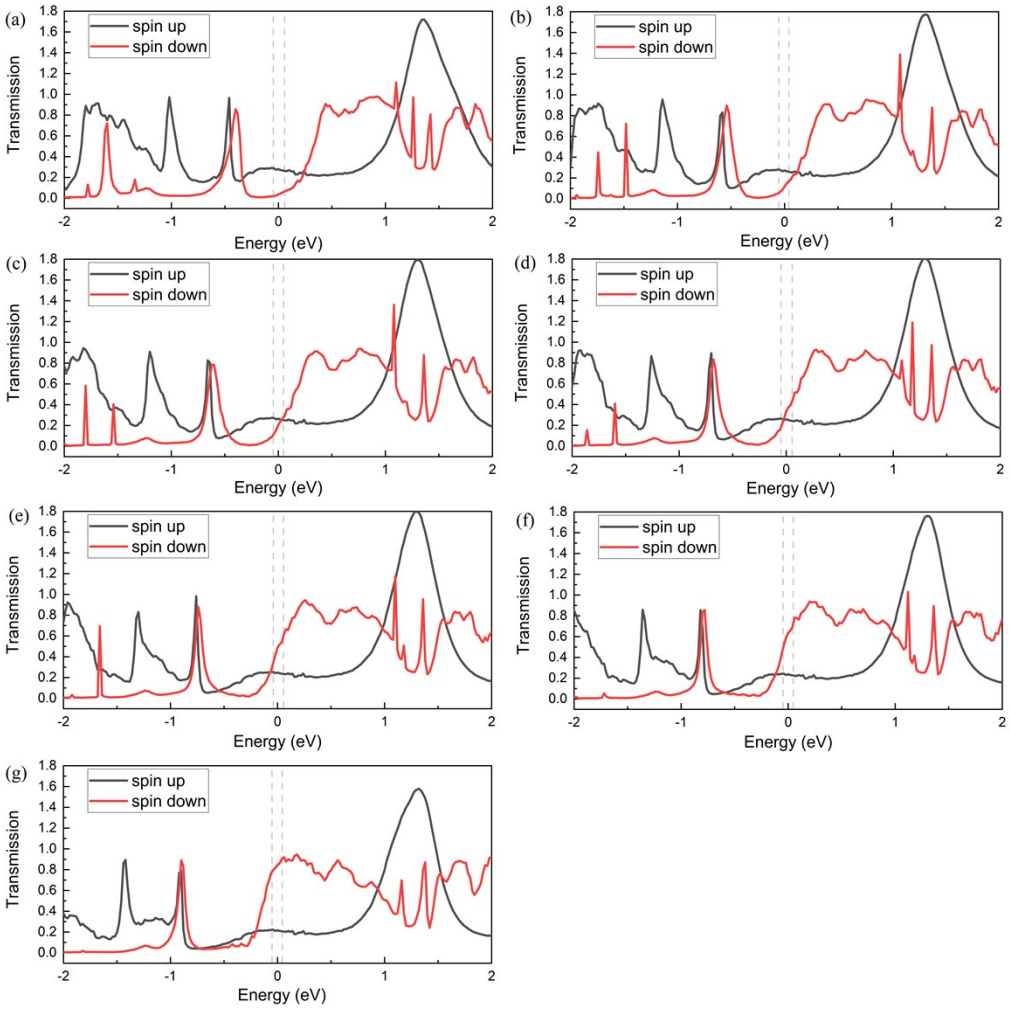

Fig. S6. Variation of transmission spectra with the gate voltage (a) -2V, (b) -1V, (c) -0.5V, (d) 0V, (e) 0.5V, (f) 1.0V, (g) 2V in C3 configuration, respectively. Here, dotted lines represent the bias voltage window of 0.1 V.

Table S1. Total energies of different spin multiplicities for isolated Mn(DBTAA) molecule.

Spin multiplicity	Energy (Hartree)
Doublet	-2064.593
Quartet	-2064.639
Sextet	-2064.605

Table S2. Magnetic exchange energy (E_{FM}-E_{AFM}, meV) of C2 under different Hubbard values (eV) for Mn and Fe atoms.

Mn \ Fe	0	1	2
0	11.66	10.33	9.89
1	8.52	7.39	7.87
2	7.60	5.78	6.29
3	5.85	4.91	5.55
4	5.82	5.46	6.68