Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

Room-Temperature Half-Metals Induced by Chemical Surface Modification: 2D Mn_2Se_2 monolayer

Zhe Wang, Yanqiu Zheng, Ji Chen, Yun Wang, Yu Liang, Xiang Li, Fang Wu*

College of Information Science and Technology, Nanjing Forestry University, Nanjing, Jiangsu 210037, P. R. China

E-mail: fangwu@njfu.edu.cn

Table S1. Optimized lattice parameters a and b (Å), magnetic moment M(μ_B) of the Mn atom, energy difference (per Mn atom) between the FM and AFM states $\Delta E = E_{FM} - E_{AFM}$ (meV) for Mn_2Se_2 calculated using the GGA+U method with different U values. The available experimental data are listed in parentheses for comparison.

U (eV)	a, b(Å)	M(μ_B)/Mn	ΔE(meV)
1	4.207	±4.160	351
2	4.264	±4.345	258
2.3	4.282 (4.28)	±4.386(±4.38)	236(232)
2.5	4.291	±4.410	221
3	4.309	±4.461	189
4	4.347	±4.546	155
5	4.375	±4.609	113

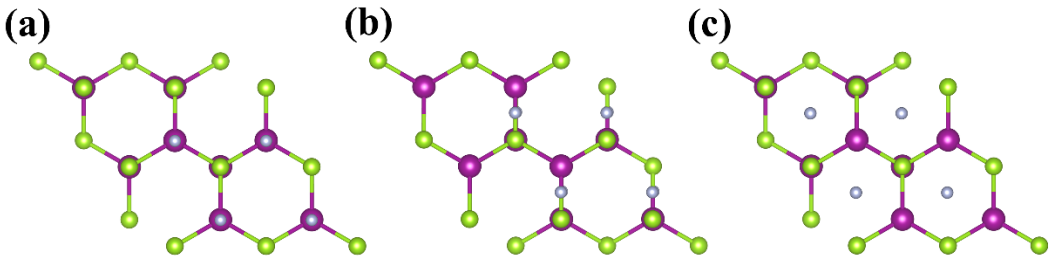

Figure S1. Schematic structures of Mn_2Se_2 monolayers modified with X atoms (X=F, Cl, Br, I and S) at the several adsorption sites (a) on-top, (b) bridge, (c) hollow. The purple, green, and gray balls represent Mn, Se, and X atoms, respectively.

Table S2. The total energies (in eV) of Mn_2Se_2 monolayer with F, Cl, Br, I and S adsorption at different adsorption sites (i.e. on-top, bridge and hollow sites).

adsorption sites	on-top	bridge	hollow
$Mn_2Se_2F_2$	-144.632	-144.388	-144.486
$Mn_2Se_2Cl_2$	-132.804	-132.548	-132.664
$Mn_2Se_2Br_2$	-128.816	-128.601	-128.716
$Mn_2Se_2I_2$	-124.936	-124.432	-123.992
$Mn_2Se_2S_2$	-137.248	-136.776	-136.410

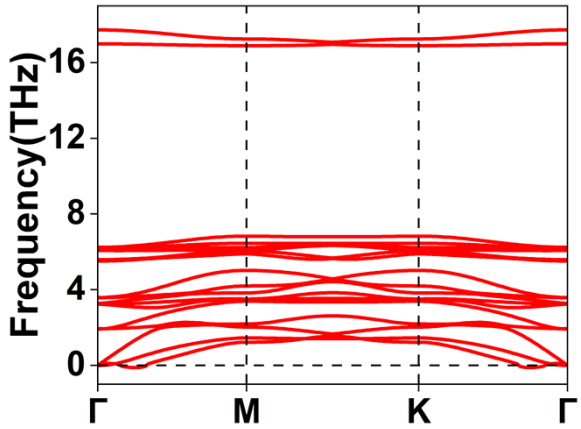

Figure S2. Phonon dispersion curves of $Mn_2Se_2F_2$ monolayer.

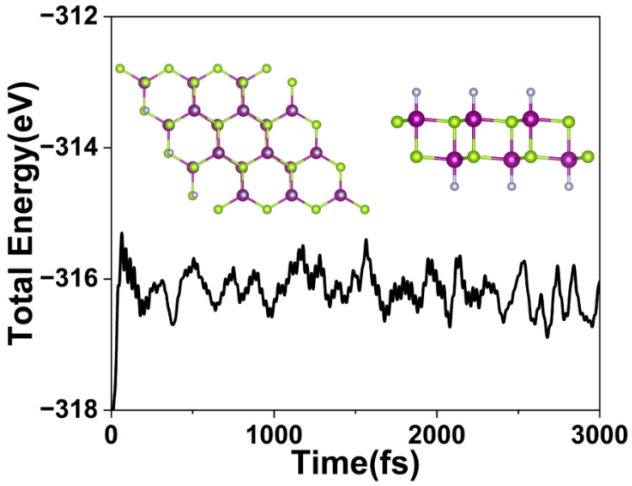

Figure S3. Evolution of total energy for the $Mn_2Se_2F_2$ monolayer at 300 K from AIMD simulations. Here, top views and side views of the corresponding geometrical structures after the AIMD simulation are shown.

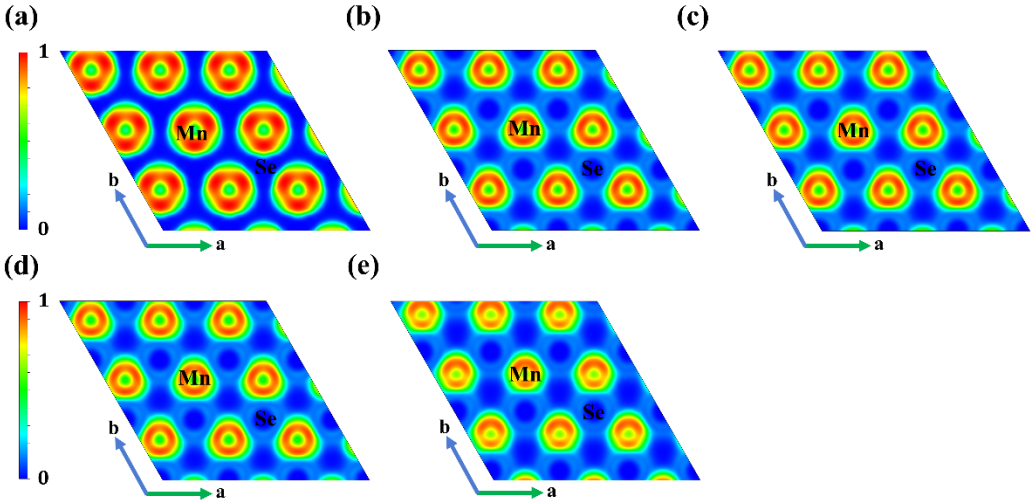

Figure S4. Electron localization functions (ELF) of (a) $Mn_2Se_2F_2$, (b) $Mn_2Se_2Cl_2$, (c) $Mn_2Se_2Br_2$, (d) $Mn_2Se_2I_2$ and (e) $Mn_2Se_2S_2$ monolayers.

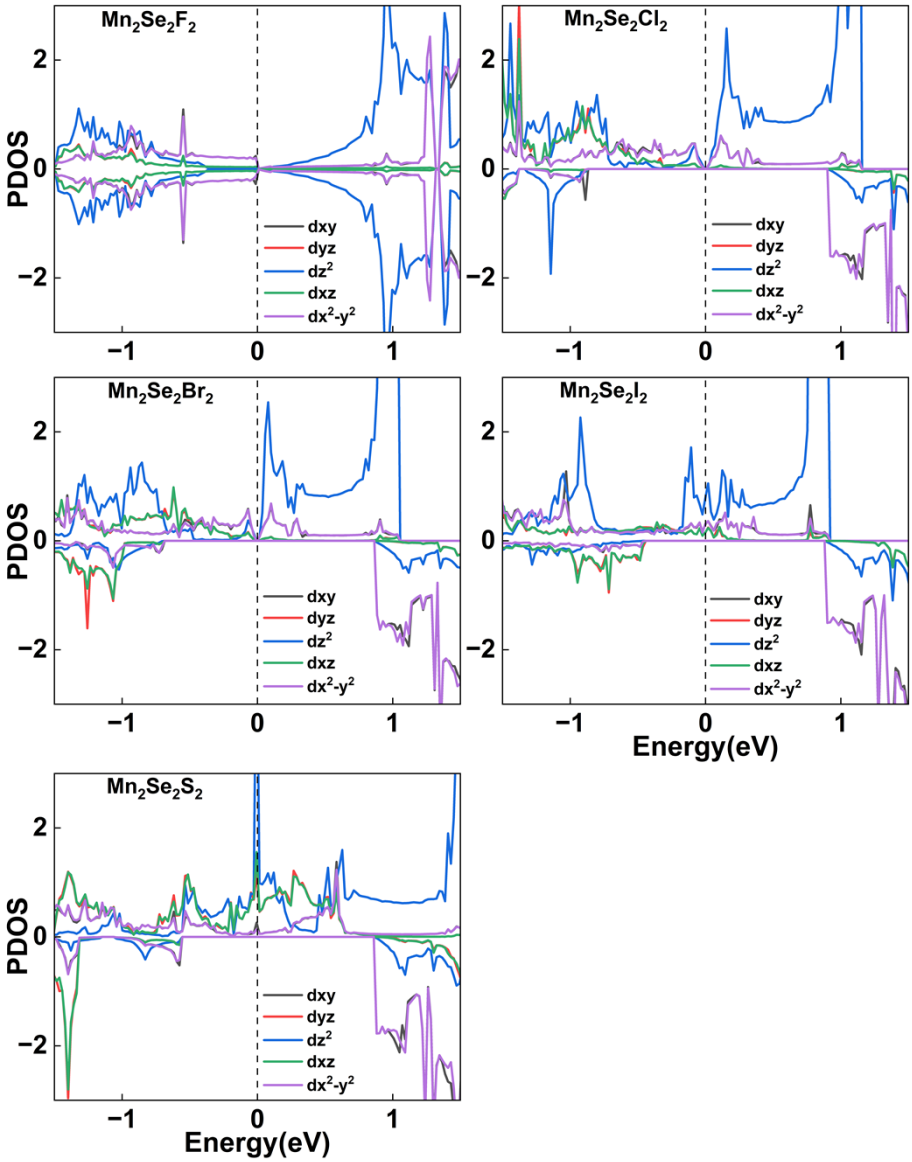

Figure S5. The PDOS of Mn d orbitals for Mn_2Se_2 derivatives monolayer.