Supporting Information

Asymmetric Janus functionalization induced magnetization and switchable out-of-plane polarization in 2D MXene Mo_2CXX'

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Figures:

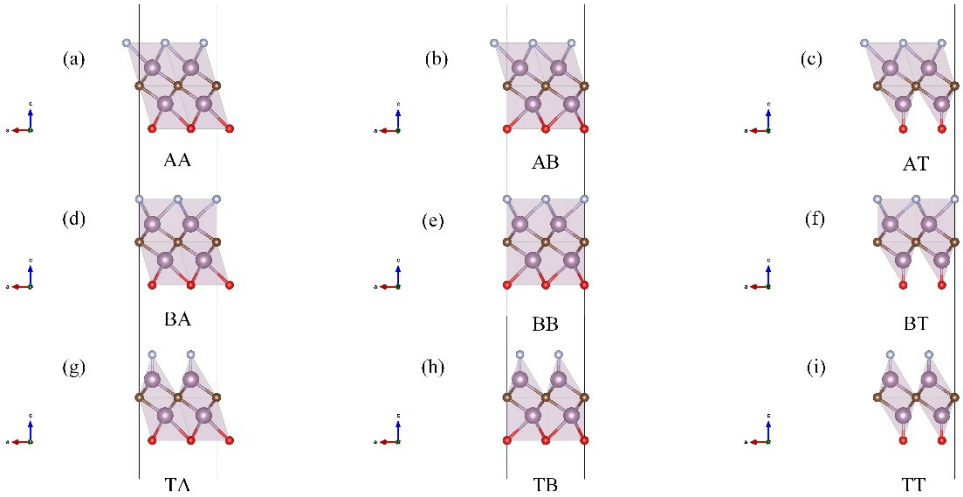

Figure ESI1. Side views of Mo_2C-FO MXene lattice. There are nine possible sites for X atoms decoration on each side of the Janus surface.

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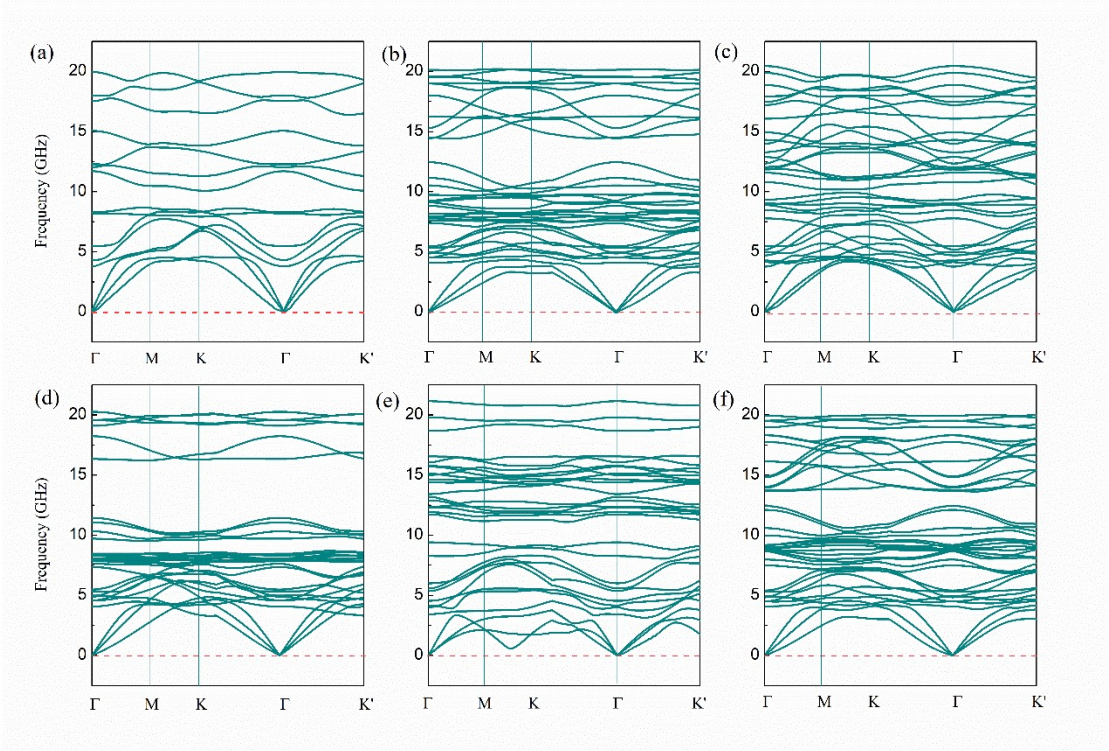

Figure ESI2. The phonon spectra for (a) Mo_2C-FO; (b) Mo_2C-F-OH; (c) Mo_2C-O-OH; (d) Mo_2C-F_2; (e) Mo_2C-O_2; (f) Mo_2C-$(OH)_2$.

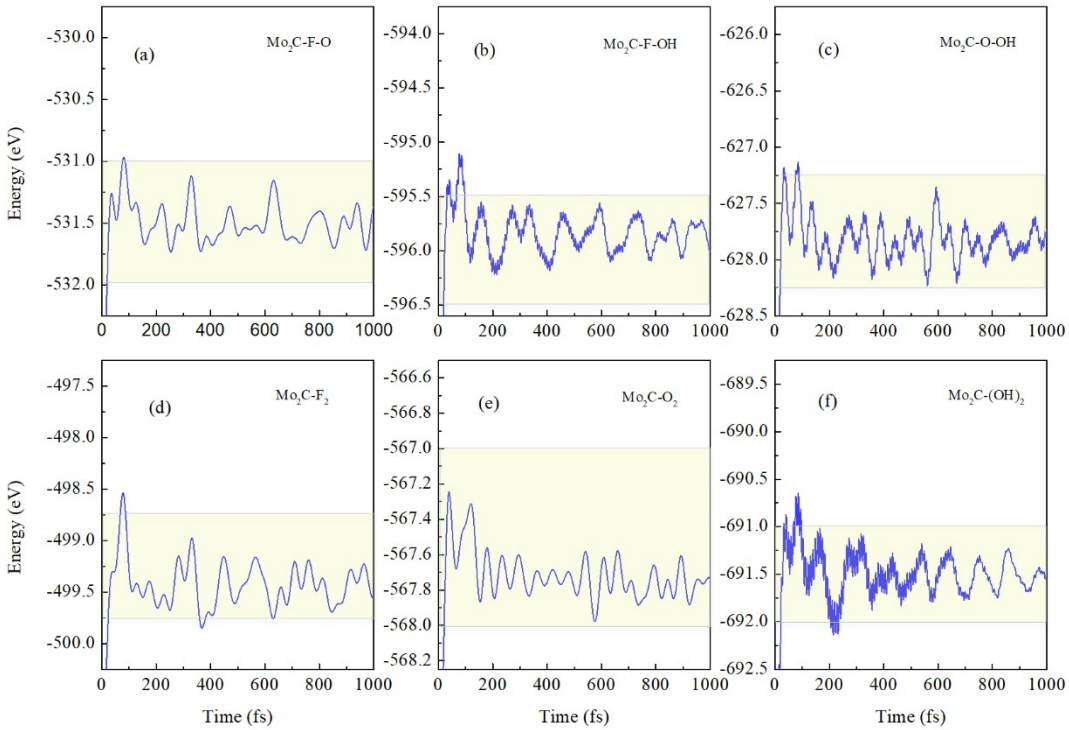

Figure ESI3. Variations of the total free energy of Janus during ab initio molecular dynamics simulations at 300 K. (a) Mo_2C-FO; (b) Mo_2C-F-OH; (c) Mo_2C-O-OH; (d) Mo_2C-F_2; (e) Mo_2C-O_2; (f) Mo_2C-$(OH)_2$.

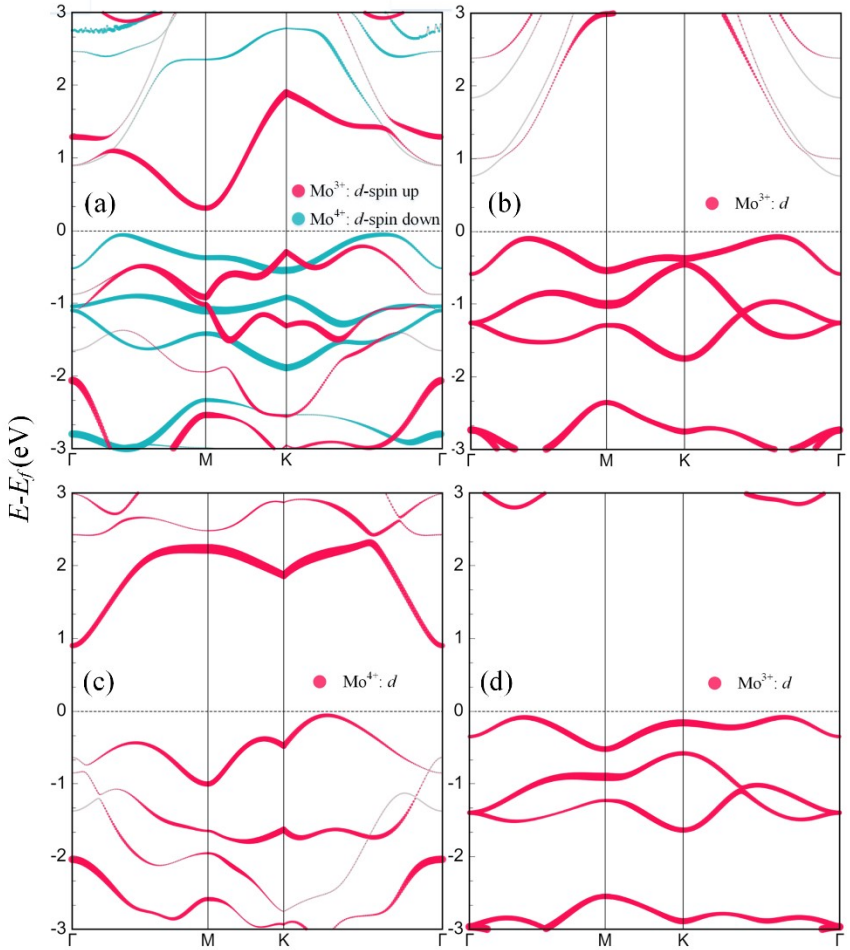

Figure ESI4. The spin polarized d orbital projected band structure of Mo ion for six functionalized Mo_2C MXenes: asymmetry Janus (a) Mo_2C-OOH, (b) Mo_2C-$(OH)_2$, (c) Mo_2C-F_2, and (d) Mo_2C-O_2 and the Fermi level defines zero energy.

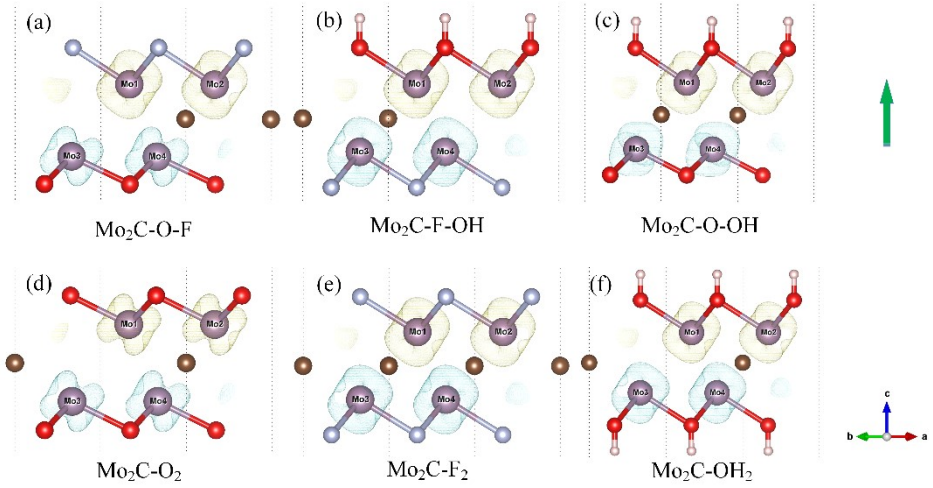

Figure ESI5. The spin density ($\rho_{\uparrow}-\rho_{\downarrow}$) map of (a) Mo_2C-FO; (b) Mo_2C-F-OH; (c) Mo_2C-O-OH; (d) Mo_2C-F_2; (e) Mo_2C-O_2; (f) Mo_2C-$(OH)_2$. The green arrow represents the direction of electric

polarization.

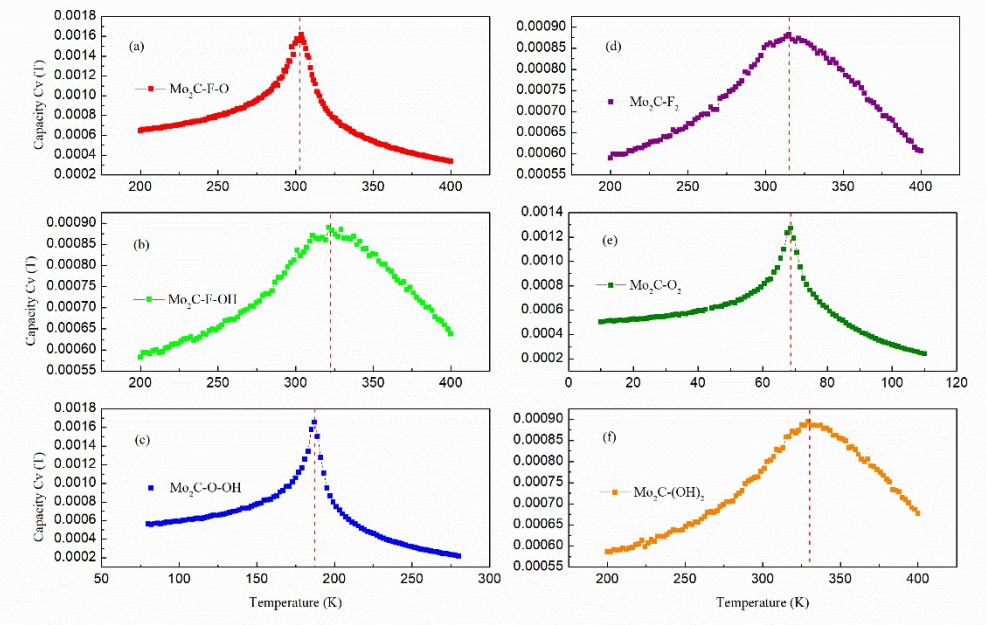

Figure ESI6. Specific heat Cv as a function of temperature for (a) Mo_2C-FO; (b) Mo_2C-F-OH; (c) Mo_2C-O-OH; (d) Mo_2C-F_2; (e) Mo_2C-O_2; (f) Mo_2C-$(OH)_2$.

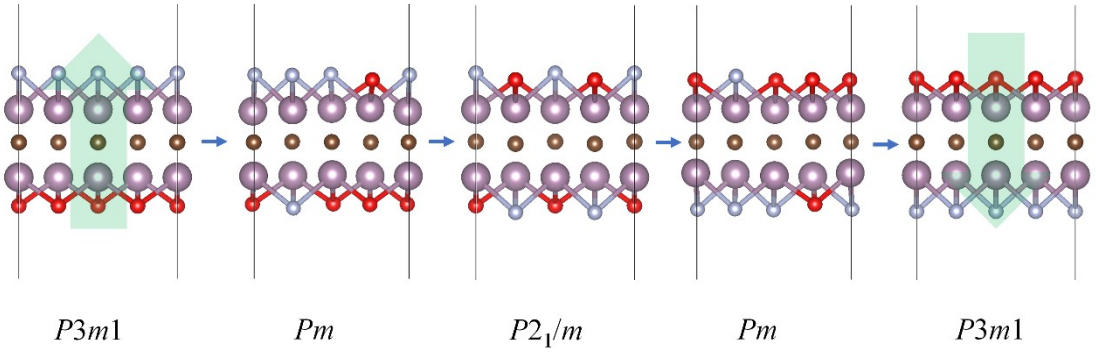

Figure ESI7. Side views of Mo_2C-FO MXene lattice. Five phases include non-centrosymmetric $P3m1$, Pm group, and centrosymmetric $P2_1/m$. The green arrow represents the direction of electric polarization.

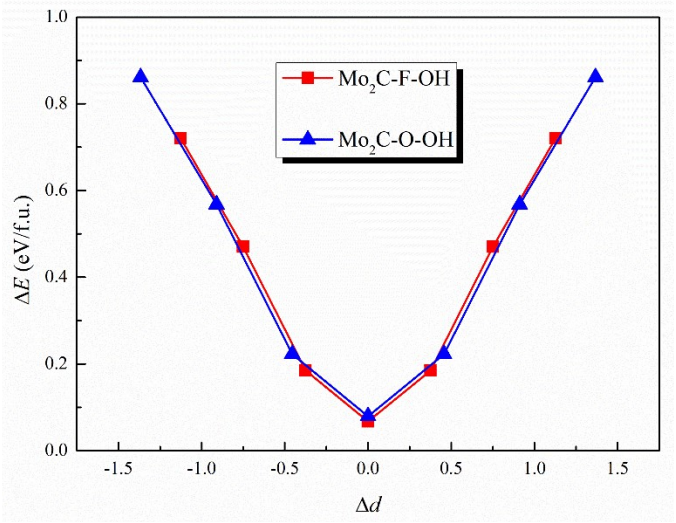

Figure ESI8. Total energy as function of the difference of layer distance Δd for Mo_2C-F-OH and Mo_2C-O-OH system.

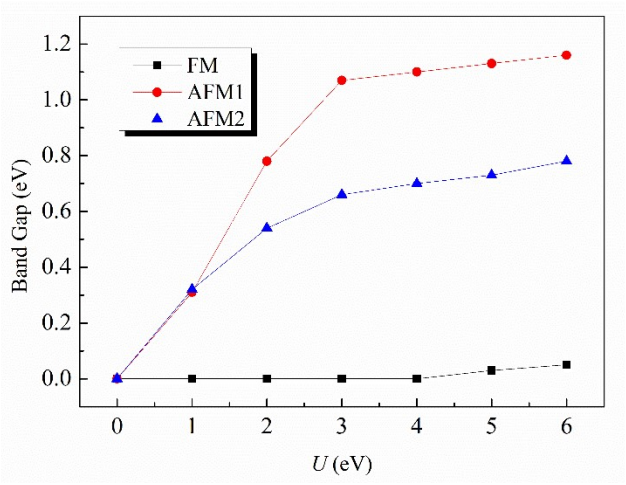

Figure ESI9. Calculated band gap with different U_{eff} values of Mo ion for three magnetic structures (FM, AFM1, AFM2).

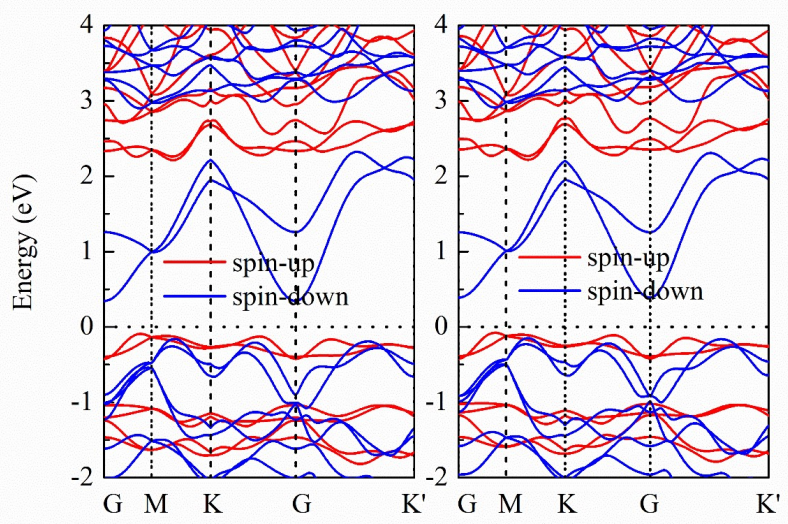

Figure ESI10. The band structure of Mo2C-O-OH calculated by (a) DFT-D2, (b) DFT-D3.

Tables

Table ESI1. Calculated total energy for three magnetic orders (FM, AFM1, AFM2) with different hubbard *U* values of Mo ion.

	U=0	*U*=1	*U*=2	*U*=3	*U*=4	*U*=5	*U*=6
FM	-91.4869	-86.5886	-81.6545	-76.7160	-72.9473	-68.6091	-65.9987
AFM1	-91.4864	-86.4594	-81.6256	-76.9326	-72.9659	-69.5811	-66.6318
AFM2	-91.4871	-86.5888	-81.7033	-76.8244	-72.7801	-69.3298	-66.4711

Table ESI2. Calculated magnetic moments for three magnetic orders (FM, AFM1, AFM2) with different hubbard *U* values of Mo ion.

	U=0	*U*=1	*U*=2	*U*=3	*U*=4	*U*=5	*U*=6
$(Mo1/Mo2)_{FM}$	0/0	0/0	0.22/0.22	0.44/1.45	2.41/-1.61	2.87/2.30	3.03/1.98
$(Mo1/Mo2)_{AFM1}$	0/0	0/0	0/0	1.32/-0.92	2.18/-1.72	2.76/-1.86	2.94/-2.03
$(Mo1/Mo2)_{AFM2}$	0/0	0/0	0.03/0	0.17/-0.17	2.15/-1.74	2.75/-1.85	2.90/-1.97

Table ESI3 Vacuum-level, level shift, and work function for six functionalized Mo_2C-MXenes.

MXenes	1Vacuum-Level (eV)	2Vacuum-Level (eV)	level shift (Δ)	Work Function (eV)
Mo_2C-F-O	3.916	3.204	0.712	6.250
Mo_2C-F-OH	4.880	0.808	4.072	5.945
Mo_2C-O-OH	5.572	0.694	4.878	6.315
Mo_2C-F_2	3.259	3.259	0.000	5.847
Mo_2C-O_2	3.808	3.808	0.000	6.294
Mo_2C-OH_2	2.451	2.451	0.000	1.842

Table ESI4. Calculated lattice parameters (Å) and band gap (eV) for DFT-D2 and DFT-D3 vdw correction.

Vdw	*a*	*b*	Band gap
DFT-D2	3.29	3.19	0.36
DFT-D3	3.28	3.18	0.43