Supplementary Information: Investigation of the electronic and magnetic properties of bare and surface functionalized double ordered MXenes for spintronic devices

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1 Linear response approach

We used linear response approach formulated by Cococcioni et al.[1] to evaluate the U parameters for DTM structures.

Quantum ESPRESSO (QE) code[2] was used for the LRA calculations. To produce results with negligible error, we make sure that similar computational parameters, such as PAW PPs, are used for both VASP and QE computations.

A typical example of LRA calculations for Cr_3C_2 and $Cr_3C_2O_2$ are displayed in Fig. S1. We first determine x_0 and x which represent the non-interacting (bare) and interacting density response functions of the system with respect to localized perturbations. The parameter U_{eff} is then evaluated from the expression: $U_{eff} = (x_0^{-1} - x^{-1})$.

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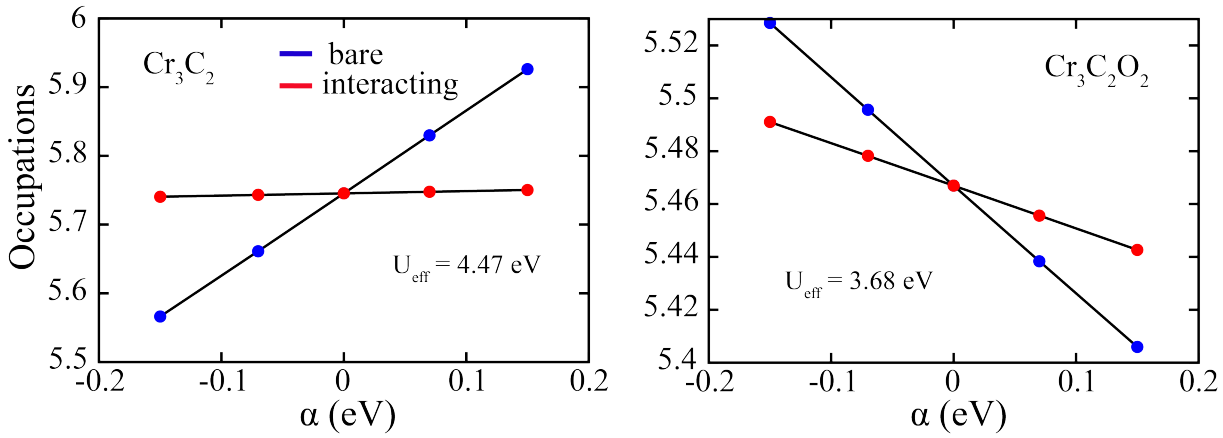

Fig. 1 Linear response of d orbital occupations as a function of potential shift α. The curves depicted by the squares and circles lines are labeled bare and interacting. The inverse response functions are deduced numerically by calculating the slope of the curves. x_0 follows from the slope of curve bare, whereas x from the slope of curve interacting.

Table 1 The calculated U_{eff} values in eV for each metal atom in bare and oxygen-terminated MXene monolayer structures.

Mxene	Ti	V	Cr	Zr	Nb	Mo	Ta
Cr_3C_2			4.71				
Ti_3C_2	3.06						
Zr_3C_2				2.06			
Cr_2NbC_2			3.67		3.30		
Cr_2TaC_2			3.97				4.06
Cr_2TiC_2	4.07		5.11				
Cr_2VC_2		4.40	4.26				
Mo_2NbC_2					4.12	4.11	
Mo_2TaC_2						4.10	4.14
Mo_2TiC_2	4.06					4.00	
Mo_2VC_2		4.06				4.14	
Ti_2NbC_2	3.41				3.47		
Ti_2TaC_2	3.41						3.71
$Cr_3C_2O_2$			3.68				
$Ti_3C_2O_2$	3.13						
$Zr_3C_2O_2$				1.81			
$Cr_2NbC_2O_2$			3.98		3.13		
$Cr_2TaC_2O_2$			3.98				2.72
$Cr_2TiC_2O_2$	3.67		5.43				
$Cr_2VC_2O_2$		4.73	4.33				
$Mo_2NbC_2O_2$					3.25	4.02	
$Mo_2TaC_2O_2$						4.02	2.88
$Mo_2TiC_2O_2$	3.46					4.00	
$Mo_2VC_2O_2$		4.48				4.09	
$Ti_2NbC_2O_2$	3.39				3.06		
$Ti_2TaC_2O_2$	3.40						2.67

Table 2 The calculated adsorption energy values (eV) for per oxygen atom on MXenes structures are tabulated with the favorable adsorption site.

	$Cr_3C_2O_2$	$Ti_3C_2O_2$	$Zr_3C_2O_2$	$Cr_2NbC_2O_2$	$Cr_2TaC_2O_2$	$Cr_2TiC_2O_2$	$Cr_2VC_2O_2$	$Mo_2NbC_2O_2$	$Mo_2TaC_2O_2$	$Mo_2TiC_2O_2$	$Mo_2VC_2O_2$	$Ti_2NbC_2O_2$	$Ti_2TaC_2O_2$
E_{ads}	-6.61	-8.17	-8.71	-6.46	-6.57	-6.60	-6.60	-6.97	-7.01	-7.27	-7.12	-8.13	-8.23
Ads. Site	C	C	C	C	C	B	C	B	B	B	B	C	C

Table 3 The optimized lattice constants $a = b$ (Å) of the bare oxygen terminated MXenes

MXene	Cr_3C_2	Ti_3C_2	Zr_3C_2	Cr_2NbC_2	Cr_2TaC_2	Cr_2TiC_2	Cr_2VC_2	Mo_2NbC_2	Mo_2TaC_2	Mo_2TiC_2	Mo_2VC_2	Ti_2NbC_2	Ti_2TaC_2
wo-U	3.025	3.097	3.344	3.121	3.102	3.078	3.006	3.043	3.026	2.960	2.940	3.139	3.123
w-U	3.162	3.128	3.371	3.148	3.196	3.153	3.087	3.161	3.163	3.084	3.032	3.163	3.152
MXene	$Cr_3C_2O_2$	$Ti_3C_2O_2$	$Zr_3C_2O_2$	$Cr_2NbC_2O_2$	$Cr_2TaC_2O_2$	$Cr_2TiC_2O_2$	$Cr_2VC_2O_2$	$Mo_2NbC_2O_2$	$Mo_2TaC_2O_2$	$Mo_2TiC_2O_2$	$Mo_2VC_2O_2$	$Ti_2NbC_2O_2$	$Ti_2TaC_2O_2$
wo-U	2.954	3.013	3.278	3.114	3.107	3.099	3.011	3.002	3.003	2.842	2.911	3.004	3.061
w-U	2.954	3.084	3.316	3.114	3.106	3.078	3.010	2.298	2.974	2.963	2.927	3.108	3.103

Table 4 The calculated cohesive (E_{coh}) and formation (E_f) energy vaules in eV/atom for bare monolayer MXene structures.

MXene	Cr_3C_2	Ti_3C_2	Zr_3C_2	Cr_2NbC_2	Cr_2TaC_2	Cr_2TiC_2	Cr_2VC_2	Mo_2NbC_2	Mo_2TaC_2	Mo_2TiC_2	Mo_2VC_2	Ti_2NbC_2	Ti_2TaC_2
E_{coh}	5.419	7.167	7.731	6.339	6.648	6.079	6.024	7.272	7.601	7.078	6.986	7.447	7.759
E_f	0.242	-0.352	-0.331	0.053	0.118	-0.033	0.108	0.296	0.341	0.144	0.322	-0.286	-0.225

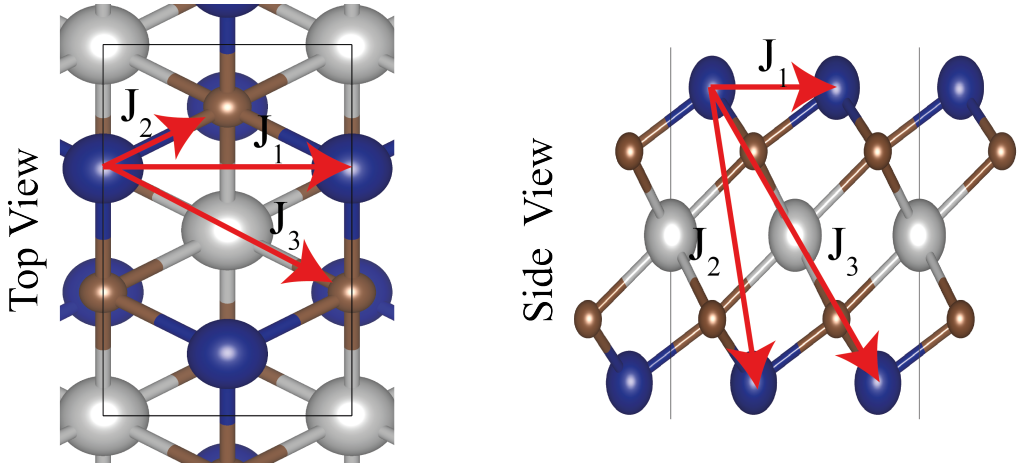

Fig. 2 Top and side views of the MXene structures with the rectangular cells. The nearest-, next-nearest- and next-next-nearest-neighbor exchange coupling parameters (J_1, J_2 and J_3, respectively) shown by arrows.

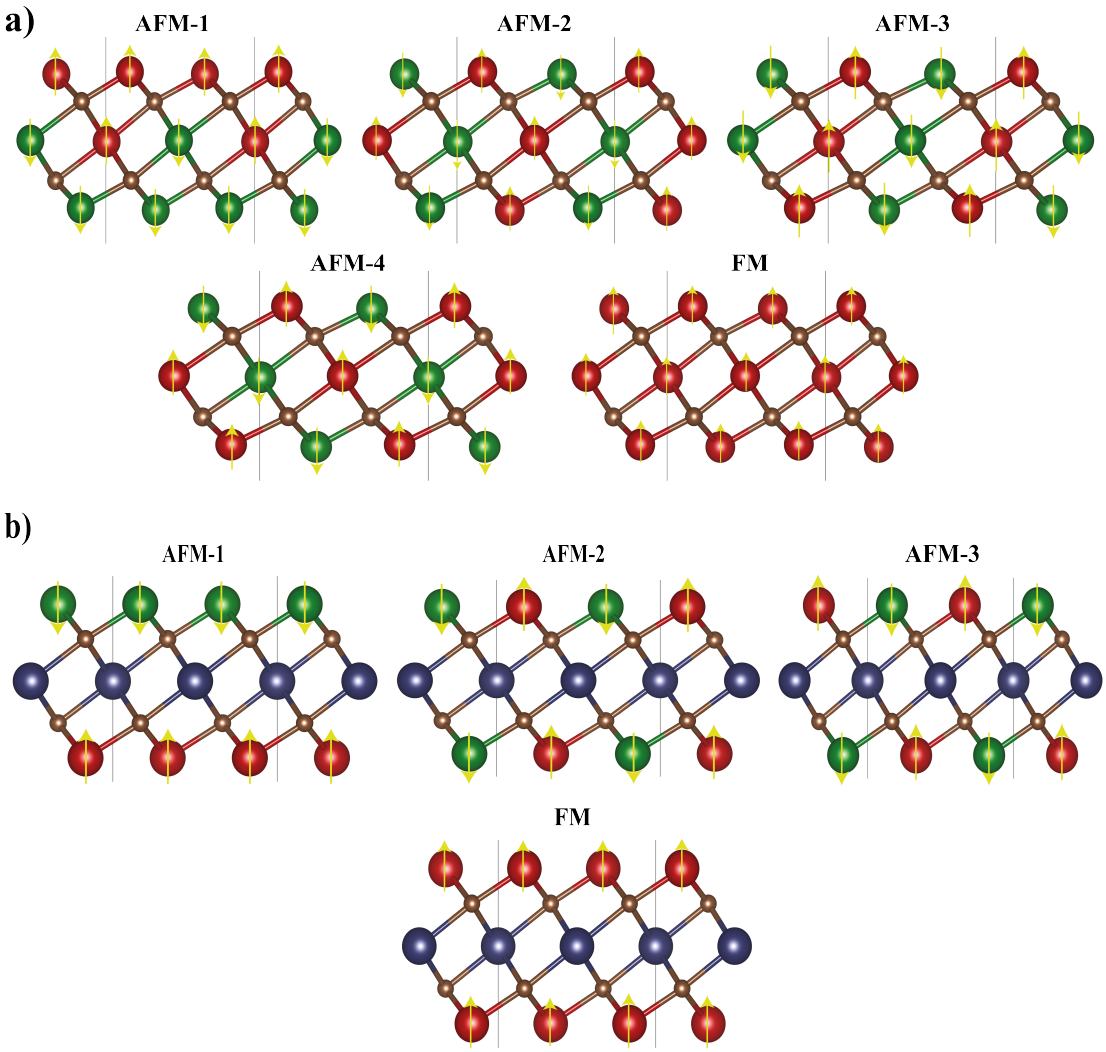

Fig. 3 Magnetic orientations for a) each metal layer atoms have magnetic moment b) middle metal layer atoms have no magnetic moment. Red and green balls illustrate spin-up and spin-down orientation, respectively.

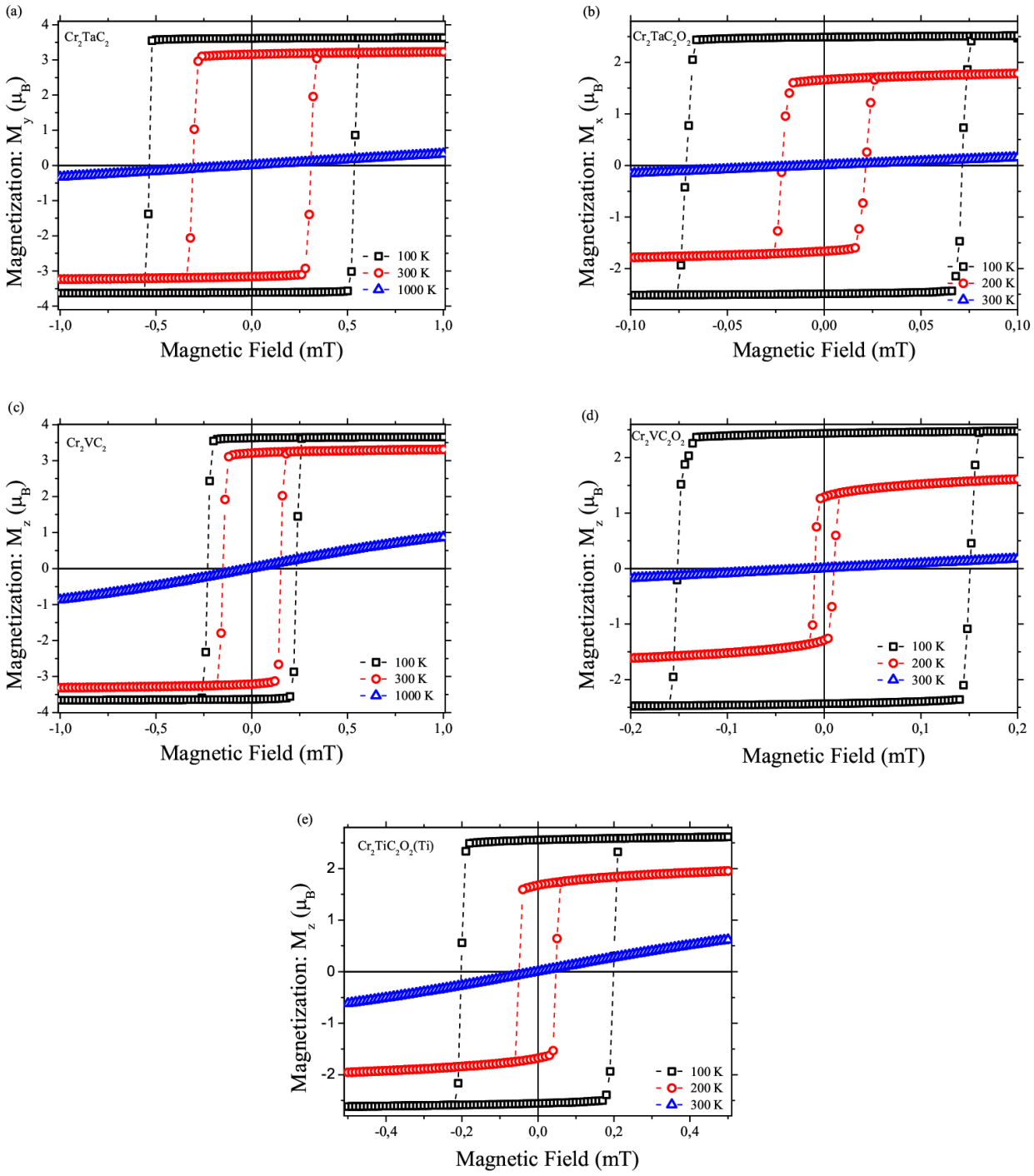

Fig. 4 Hysteresis curves of Cr-based MXene structures at several temperatures. .

Table 5 Total energy value (in unit of eV) per formula unit for different magnetic orientations.

MXene	AFM1	AFM2	AFM3	FM	
Cr_2NbC_2	-39.93363466	-39.89614783	-39.79280551	-40.28894902	
$Cr_2NbC_2O_2$	-55.82777373	-55.73574424	-55.73539916	-55.84864899	
Cr_2TiC_2	-35.62423928	-34.8499153	-34.86805635	-35.58448503	
$Cr_2TiC_2O_2$ (C)	-49.17107243	-49.18977845	-49.20004705	-49.17655127	
$Cr_2TiC_2O_2$ (Ti)	-50.54057159	-50.41220599	-50.424833	-50.54418349	
Cr_2VC_2	-35.55066893	-35.13583162	-35.10488067	-35.58362736	
$Cr_2VC_2O_2$	-51.02206546	-50.9494969	-50.95343245	-51.05433624	
Cr_2TaC_2	-40.11820197	-40.16784456	-40.10189687	-40.42710694	
$Cr_2TaC_2O_2$	-57.86943942	-57.78049557	-57.77948186	-57.89734561	
Ti_3C_2	-36.539260006	-36.277446085	-36.293154265	-36.43362736	
$Ti_3C_2O_2$	-56.04564962	-56.045656865	-56.045654085	-56.045646315	
Zr_3C_2	-42.29698757	-42.19103438	-42.18166457	-42.25272169	
$Zr_3C_2O_2$	-63.84851385	-63.84851472	-63.84851494	-63.84851332	
Mo_2NbC_2	-37.773355645	-38.208717855	-38.197761875	-38.078943085	
Mo_2TaC_2	-38.713913385	-39.275342075	-39.239666205	-38.72370495	
Mo_2TiC_2	-	-35.49640249	-35.45696606	-35.24654412	
$Mo_2TiC_2O_2$	-54.028703125	-54.028698795	-54.02869796	-54.02870732	
Mo_2VC_2	-	-35.06399216	-35.12091152	-34.890166165	
Ti_2NbC_2	-38.236896235	-38.158908405	-38.171593025	-38.24506252	
$Ti_2NbC_2O_2$	-58.199017165	-58.19900676	-58.19900241	-58.198997465	
Ti_2TaC_2	-39.091422545	-39.024745335	-39.028009125	-38.790671305	
$Ti_2TaC_2O_2$	-60.030715305	-60.030754905	-60.030730815	-60.0307384	
MXene	AFM1	AFM2	AFM3	AFM4	FM
Cr_3C_2	-35.49828528	-34.77116233	-34.88776263	-34.59715347	-35.66359075
$Cr_3C_2O_2$	-52.56146121	-52.43882712	-52.52808045	-52.37321699	-52.73552186

Table 6 Total energy value (in unit of eV) per formula unit for the energetically most stable magnetic orientations (including SOC).

MAE (eV)	001	010	100
Cr_3C_2	-34.27745623	-34.27745248	-34.277419025
$Cr_3C_2O_2$	-52.743875	-52.7437452	-52.7437375
Ti_3C_2	-36.543246	-36.5432155	-36.543217
$Ti_3C_2O_2$	-	-	-
Zr_3C_2	-42.3511375	-42.3507555	-42.350753
$Zr_3C_2O_2$	-	-	-
Cr_2NbC_2	-38.954550355	-38.95470851	-38.95418005
$Cr_2NbC_2O_2$	-53.84957	-53.84938	-53.849615
Cr_2TaC_2	-38.72029892	-38.721267595	-38.721212845
$Cr_2TaC_2O_2$	-58.02628	-58.026137	-58.02639
Cr_2TiC_2	-34.379664305	-34.37962562	-34,37962554
$Cr_2TiC_2O_2$(C)	-49.2059305	-49.2060275	-49.2060485
$Cr_2TiC_2O_2$(Ti)	-50.550565	-50.550495	-50.550505
Cr_2VC_2	-34.356312285	-34.356284255	-34.35628311
$Cr_2VC_2O_2$	-51.060775	-51.060725	-51.06072
Mo_2NbC_2	-36.48127681	-36.480902455	-36.480828575
$Mo_2NbC_2O_2$	-	-	-
Mo_2TaC_2	-37.113683915	-37.11336528	-37.11384288
$Mo_2TaC_2O_2$	-	-	-
Mo_2TiC_2	-33.92451676	-33.924292	-33.924938665
$Mo_2TiC_2O_2$	-	-	-
Mo_2VC_2	-33.55538638	-33.55522778	-33.555568425
$Mo_2VC_2O_2$	-	-	-
Ti_2NbC_2	-36.8831069	-	-
$Ti_2NbC_2O_2$	-	-	-
Ti_2TaC_2	-	-	-
$Ti_2TaC_2O_2$	-	-	-

Notes and references

1 M. Cococcioni, & S. De Gironcoli, Linear response approach to the calculation of the effective interaction parameters in the LDA+ U method, Physical Review B **71**, 035105 (2005).

2 P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. Chiarotti, M. Cococcioni, I. Dabo, & Others, QUANTUM ESPRESSO: a modular and open-source software project for quantum simulations of materials, Journal Of Physics: Condensed Matter **21**, 395502 (2009).