Electronic Supplementary Information to

Prediction of induced magnetism in 2D Ti_2C based MXene by manipulating the mixed surface functionalization and metal substitution computed by xTB model Hamiltonian of the DFTB method

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1 Energy difference by DFT

Density functional theory (DFT) calculations were carried out using Quantum espresso Package [1, 2]. The electronic exchange-correlation interaction was described by the functional of Perdew, Burke, and Ernzerhof (PBE) within the generalized gradient approximation (GGA) [3]. Energy and force convergence criteria of 10^{-4} a.u and 2×10^{-5} a.u, respectively, were used in all calculations, along with a kinetic energy cutoff of 80 Ry to expand the wave functions, a 6×6×1 Monkhorst-Pack k-point grid
[4] and a Marzari-Vanderbilt smearing width [5] of 0.05 Ry.

Table S1: PBE calculations of the magnetic energy difference ($E_{FM,AFMj(j=1,2,3)} - E_{F\ M}$) in meV and the corresponding magnetic moments in μ_B/cell of mixed $Ti_2CA_xB_y$ (A, B = O, F, OH). The ground state energy is marked in bold.

	Magnetic energy difference				Total magnetic moments			
Initial magnetic state	FM	AFM1	AFM2	AFM3	FM	AFM1	AFM2	AFM3
$Ti_2CO_1(OH)_1$	**0.00**	16.30	8.10	8.10	**1.05**	1.00	0.07	0.02
$Ti_2CO_{1.5}(OH)_{0.5}$	**0.00**	0.00	0.00	0.00	**0.97**	0.97	0.97	0.97
$Ti_2CO_{0.5}(OH)_{1.5}$	0.00	-10.80	**-12.20**	-2.70	1.69	0.36	**1.31**	0.05
$Ti_2CO_1F_1$	**0.00**	0.00	38.10	38.10	**1.96**	1.96	0.01	0.02
$Ti_2CO_{1.5}F_{0.5}$	**0.00**	0.00	0.00	0.00	**1.00**	1.00	1.00	1.00
$Ti_2CO_{0.5}F_{1.5}$	**0.00**	80.30	32.60	5.40	**2.92**	0.60	1.82	0.88
$Ti_2CF_1(OH)_1$	0.00	34.00	**-59.80**	-34.00	2.80	0.72	**0.00 (AFM2)**	0.00 (AFM3)
$Ti_2CF_{1.5}(OH)_{0.5}$	0.00	79.00	**-27.20**	69.40	3.85	0.23	**0.00 (AFM2)**	0.16
$Ti_2CF_{0.5}(OH)_{1.5}$	**0.00**	47.60	95.20	39.30	**3.31**	0.94	0.63	0.50

2 Substitution of Ti by Sc in mixed surface termination ($Ti_2CA_xB_y$).

Table S2: Absolute value of the local magnetic moments (in μ_B) in $Ti_2CA_xB_y$.

	Ti	C
$Ti_2CO_{1.0}(OH)_{1.0}$	0.40 to 1.13	0.41 to 0.50
$Ti_2CO_{1.5}(OH)_{0.5}$	0.02 to 1.13	0.23 to 0.27
$Ti_2CO_{0.5}(OH)_{1.5}$	0.41 to 2.00	0.27 to 0.43
$Ti_2CO_{1.0}F_{1.0}$	0.51 to 0.96	0.34 to 0.40
$Ti_2CO_{1.5}F_{0.5}$	0.00 to 1.01	0.18 to 0.22
$Ti_2CO_{0.5}F_{1.5}$	0.40 to 1.45	0.44 to 0.46
$Ti_2CF_{1.0}(OH)_{1.0}$	1.50 to 1.70	0.00
$Ti_2CF_{1.5}(OH)_{0.5}$	1.40 to 1.70	0.00
$Ti_2CF_{0.5}(OH)_{1.5}$	1.26 to 1.60	0.71 to 0.74

Table S3: Absolute value of the local magnetic moments (in μ_B) in Ti substituted by Sc in $Ti_2CA_xB_y$.

	Ti	C	Sc
Sc in $Ti_2CO_{1.0}(OH)_{1.0}$	0.32 to 1.52	0.42 to 0.60	0.10
Sc in $Ti_2CO_{1.5}(OH)_{0.5}$	0.00 to 1.48	0.24 to 0.30	0.02
Sc in $Ti_2CO_{0.5}(OH)_{1.5}$	0.10 to 1.71	0.60 to 0.70	0.13
Sc in $Ti_2CO_{1.0}F_{1.0}$	0.47 to 1.25	0.31 to 0.43	0.08
Sc in $Ti_2CO_{1.5}F_{0.5}$	0.00 to 1.30	0.12 to 0.24	0.02
Sc in $Ti_2CO_{0.5}F_{1.5}$	0.61 to 1.53	0.45 to 0.50	0.11
Sc in $Ti_2CF_{1.0}(OH)_{1.0}$	1.28 to 1.83	0.66 to 0.75	0.18
Sc in $Ti_2CF_{1.5}(OH)_{0.5}$	1.17 to 1.71	0.60 to 0.64	0.14
Sc in $Ti_2CF_{0.5}(OH)_{1.5}$	1.10 to 1.71	0.72 to 0.79	0.20

3 Substitution of Ti by Sc in 5×5×1-Ti_2CO_2

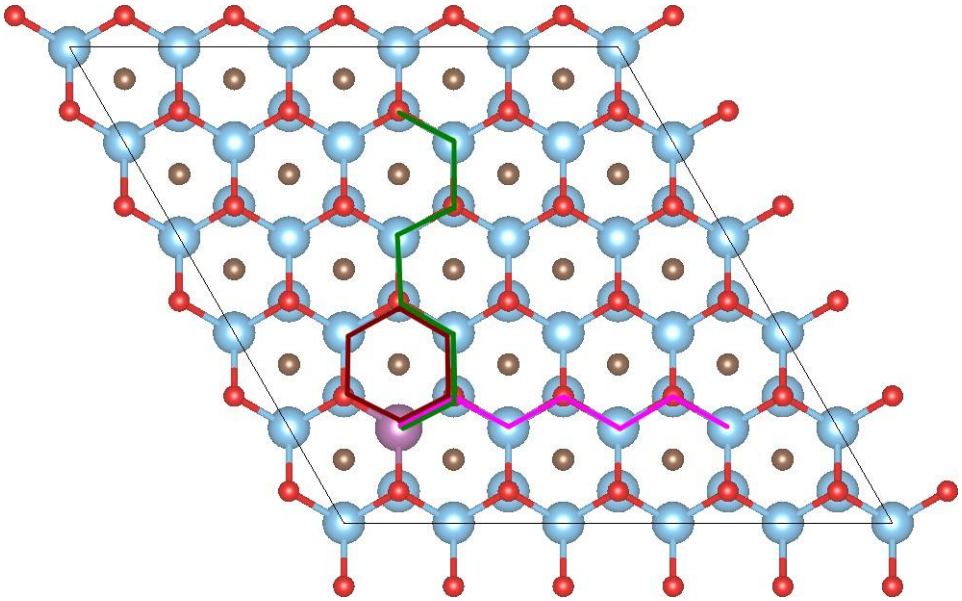

Figure S1: Structure of Ti substituted by Sc in 5×5×1-Ti_2CO_2 following the Zigzag, Armchair and Cluster directions.

Table S4: Total energies of n Ti atoms substituted by n Sc atoms in 5×5×1-Ti_2CO_2 following the Zigzag, Armchair, and Cluster directions. 1, 2, and 3 Sc in the cell have the same positions for all the considered directions, and 4Sc atoms following the cluster are the same as in the armchair direction (See Figure S1).

	Armchair	Zigzag	Cluster
1Sc	-9637.2446	–	–
2Sc	-9630.9588	–	–
3Sc	-9624.3116	–	–
4Sc	-9617.4778	-9617.6027	–
5Sc	-9610.3680	-9610.3667	-9610.2819
6Sc	-9603.1222	-9603.1358	-9602.8932
7Sc	-9596.1266	-9595.8059	–

References

[1] P Giannozzi, S Baroni, N Bonini, M Calandra, R Car, C Cavazzoni, D Ceresoli, G L Chiarotti, M Cococcioni, I Dabo, A Dal Corso, S de Gironcoli, S Fabris, G Fratesi, R Gebauer, U Gerstmann, C Gougoussis, A Kokalj, M Lazzeri, L Martin-Samos, N Marzari, F Mauri, R Mazzarello, S Paolini, A Pasquarello, L Paulatto, C Sbraccia, S Scandolo, G Sclauzero, A P Seitsonen, A Smogunov, P Umari, and R M Wentzcovitch. Quantum espresso: a modular and open-source software project for quantum simulations of materials. *Journal of Physics: Condensed Matter*, 21:395502, 2009.

[2] P Giannozzi, O Andreussi, T Brumme, O Bunau, M Buongiorno Nardelli, M Calandra, R Car, C Cavazzoni, D Ceresoli, M Cococcioni, N Colonna, I Carnimeo, A Dal Corso, S de Gironcoli, P Delugas, R A DiStasio, A Ferretti, A Floris, G Fratesi, G Fugallo, R Gebauer, U Gerstmann, F Giustino, T Gorni, J Jia, M Kawamura, H-Y Ko, A Kokalj, E Küçükbenli, M Lazzeri, M Marsili, N Marzari, F Mauri, N L Nguyen, H-V Nguyen, A Otero de-la Roza, L Paulatto, S Poncé, D Rocca, R Sabatini, B Santra, M Schlipf, A P Seitsonen, A Smogunov, I Timrov, T Thonhauser, P Umari, N Vast, X Wu, and S Baroni. Advanced capabilities for materials modelling with quantum espresso. *Journal of Physics: Condensed Matter*, 29:465901, 2017.

[3] J P Perdew, K Burke, and M Ernzerhof. Generalized gradient approximation made simple. *Phys. Rev. Lett.*, 77:3865–3868, 1996.

[4] H J Monkhorst and J D Pack. Special points for brillouin-zone integrations. *Phys. Rev. B*, 13:5188– 5192, 1976.

[5] P L Silvestrelli, N Marzari, D Vanderbilt, and M Parrinello. Maximally-localized wannier functions for disordered systems: Application to amorphous silicon. *Solid State Communications*, 107:7–11, 1998.