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Supplementary Materials

## **Theoretical Study on the Electronic Structure and Magnetic Properties Regulation of Janus Structure of M'MCO<sub>2</sub> 2D MXenes**

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## Basic information of lattice parameters after structural optimization

Table S1 the lattice parameters of VCrCO<sub>2</sub>

	Initial	NM	FM	AFM1	AFM2	AFM3
Symmetry	P3m1	P3m1	P3m1	Cm	P3m1	P3m1
Group	(C <sub>3V-1</sub> )	(C <sub>3V-1</sub> )	(C <sub>3V-1</sub> )	(C <sub>s-3</sub> )	(C <sub>3V-1</sub> )	(C <sub>3V-1</sub> )
IT Number	156	156	156	8	156	156
a/Å	2.88	2.85	2.95	2.96	5.87	5.88
b/Å	2.88	2.85	2.95	2.96	2.95	2.96
c/Å	21.81	21.81	21.81	21.81	21.81	21.81
$\alpha$	90.00°	90.00°	90.00°	90.00°	90.00°	90.00°
$\beta$	90.00°	90.00°	90.00°	90.00°	90.00°	90.00°
$\gamma$	120.00°	120.00°	120.00°	119.73°	120.24°	120.25°

Table S2 the lattice parameters of VMnCO<sub>2</sub>

	Initial	NM	FM	AFM1	AFM2	AFM3
Symmetry	P3m1	P3m1	Cm	Cm	Pc	Cm
Group	(C <sub>3V-1</sub> )	(C <sub>3V-1</sub> )	(C <sub>s-3</sub> )	(C <sub>s-3</sub> )	(C <sub>s-2</sub> )	(C <sub>s-3</sub> )
IT Number	156	156	8	8	7	8
a/Å	2.89	2.87	2.96	3.01	5.92	5.90
b/Å	2.89	2.87	2.96	2.97	2.96	2.99
c/Å	21.81	21.81	21.81	21.81	21.81	21.81
$\alpha$	90.00°	90.00°	90.00°	90.00°	90.00°	90.00°
$\beta$	90.00°	90.00°	90.00°	90.00°	90.00°	90.00°
$\gamma$	120.00°	120.00°	119.41°	120.55°	120.24°	120.40°

Table S3 the lattice parameters of CrMnCO<sub>2</sub>

	Initial	NM	FM	AFM1	AFM2	AFM3
Symmetry	P3m1	P3m1	P3m1	P3m1	P3m1	Cm
Group	(C <sub>3v-1</sub> )	(C <sub>3v-1</sub> )	(C <sub>3v-1</sub> )	(C <sub>3v-1</sub> )	(C <sub>3v-1</sub> )	(C <sub>s-3</sub> )
IT Number	156	156	156	156	156	8
a/Å	2.66	2.66	2.93	2.88	5.76	6.61
b/Å	2.66	2.66	2.93	2.88	2.91	3.33
c/Å	21.81	21.81	21.81	21.81	21.81	21.81
$\alpha$	90.00°	90.00°	90.00°	90.00°	90.00°	90.00°
$\beta$	90.00°	90.00°	90.00°	90.00°	90.00°	90.00°
$\gamma$	120.00°	120.00°	120.00°	120.00°	120.41°	120.37°

Table S4 the lattice parameters of V<sub>2</sub>CO<sub>2</sub>

	Initial	NM	FM	AFM1	AFM2	AFM3
Symmetry	P $\bar{3}$ m1	P $\bar{3}$ m1	C2/m	C2/m (C <sub>2h-</sub>	C2/m	P2/c (C <sub>2h-</sub>
Group	(D <sub>3d-3</sub> )	(D <sub>3d-3</sub> )	(C <sub>2h-3</sub> )	3)	(C <sub>2h-3</sub> )	4)
IT Number	164	164	12	12	12	13
a/Å	2.88	2.88	2.95	3.06	5.98	5.99
b/Å	2.88	2.88	2.95	2.96	2.94	2.95
c/Å	21.81	21.81	21.81	21.81	21.81	21.81
$\alpha$	90.00°	90.00°	90.00°	90.00°	90.00°	90.00°
$\beta$	90.00°	90.00°	90.00°	90.00°	90.00°	90.00°
$\gamma$	120.00°	120.00°	117.78°	121.17°	119.47°	119.52°

Table S5 the lattice parameters of Cr<sub>2</sub>CO<sub>2</sub>

	Initial	NM	FM	AFM1	AFM2	AFM3
Symmetry	$P\bar{3}m1$	$P\bar{3}m1$	$P\bar{3}m1$	$P\bar{3}m1$	$P\bar{3}m1$	C2/m
Group	(D <sub>3d-3</sub> )	(D <sub>3d-3</sub> )	(D <sub>3d-3</sub> )	(D <sub>3d-3</sub> )	(D <sub>3d-3</sub> )	(C <sub>2h-3</sub> )
IT Number	164	164	164	164	164	12
a/Å	2.68	2.68	2.89	2.76	5.64	5.87
b/Å	2.68	2.68	2.89	2.76	2.83	2.88
c/Å	21.81	21.81	21.81	21.81	21.81	21.81
$\alpha$	90.00°	90.00°	90.00°	90.00°	90.00°	90.00°
$\beta$	90.00°	90.00°	90.00°	90.00°	90.00°	90.00°
$\gamma$	120.00°	120.00°	120.00°	120.05°	120.08°	119.43°

Table S6 the lattice parameters of Mn<sub>2</sub>CO<sub>2</sub>

	Initial	NM	FM	AFM1	AFM2	AFM3
Symmetry	$P\bar{3}m1$	$P\bar{3}m1$	$P\bar{3}m1$	$P\bar{3}m1$	$P\bar{3}m1$	$P\bar{3}m1$
Group	(D <sub>3d-3</sub> )	(D <sub>3d-3</sub> )	(D <sub>3d-3</sub> )	(D <sub>3d-3</sub> )	(D <sub>3d-3</sub> )	(D <sub>3d-3</sub> )
IT Number	164	164	164	164	164	164
a/Å	2.87	2.73	2.95	2.97	5.85	5.87
b/Å	2.87	2.73	2.95	2.97	2.97	2.93
c/Å	21.81	21.81	21.81	21.81	21.81	21.81
$\alpha$	90.00°	90.00°	90.00°	90.00°	90.00°	90.00°
$\beta$	90.00°	90.00°	90.00°	90.00°	90.00°	90.00°
$\gamma$	120.00°	120.00°	120.00°	120.00°	120.42°	119.93°

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## Magnetic moment information

The unit of atomic magnetic moment is  $\mu\text{B}/\text{atom}$ , and the unit of total magnetic moment is  $\mu\text{B}/\text{formula unit}$ .

Table S7 the magnetic moment of  $\text{VCrCO}_2$

	FM	AFM1	AFM2	AFM3
V	0.762	0.594	0.307	0.306
Cr	2.801	2.786	2.785	2.786
C	0.259	0.018	0.015	0.015
O	0.118	0.099	0.026	0.026
total	3.068	2.124	0.001	0.003

Table S8 the magnetic moment of  $\text{VMnCO}_2$

	FM	AFM1	AFM2	AFM3
V	1.039	1.192	1.145	0.933
Mn	3.379	3.371	3.318	3.277
C	0.223	0.099	0.025	0.048
O	0.076	0.063	0.026	0.018
total	4.042	2.059	0.000	0.000

Table S9 the magnetic moment of  $\text{CrMnCO}_2$

	FM	AFM1	AFM2	AFM3
Cr	2.585	2.457	2.652	3.071
Mn	3.706	3.686	3.59	4.333
C	0.552	0.035	0.023	0.199
O	0.264	0.141	0.071	0.049
total	5.212	1.112	0.001	0.000

Table S10 the magnetic moment of  $\text{V}_2\text{CO}_2$

	FM	AFM1	AFM2	AFM3
V	1.115	1.116	1.052	0.978
C	0.136	0.000	0.000	0.042

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O	0.056	0.053	0.032	0.031
total	1.982	0.000	0.000	0.000

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Table S11 the magnetic moment of Cr<sub>2</sub>CO<sub>2</sub>

	FM	AFM1	AFM2	AFM3
Cr	2.582	2.321	2.430	2.460
C	0.516	0.003	0.024	0.270
O	0.232	0.104	0.077	0.077
total	4.184	0.034	0.386	0.019

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Table S12 the magnetic moment of Mn<sub>2</sub>CO<sub>2</sub>

	FM	AFM1	AFM2	AFM3
Mn	3.332	3.413	3.385	3.146
C	0.311	0.000	0.000	0.073
O	0.127	0.054	0.022	0.031
total	6.097	0.000	0.000	0.000

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