

**The Nature of the Electronic Ground State of M_2C (M= Ti, V, Cr, Zr, Nb,
Mo, Hf, Ta, and W) MXenes**

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Table S1. Calculated HSE06 and PBE (in parenthesis) Bader charges (Q in a.u.) of metal and carbon atoms in M_2C (M=Ti, V, Cr, Zr, Nb, Mo, Hf, Ta, and W) using plane waves.

MXene	Q(C)	Q(M)
Ti₂C	-2.62 (-2.35)	1.31 (1.17)
Zr₂C	-2.44 (-2.16)	1.22 (1.08)
Hf₂C	-2.75 (-2.47)	1.37 (1.24)
V₂C	-2.22 (-2.04)	1.11 (1.02)
Nb₂C	-2.12 (1.92)	1.06 (0.96)
Ta₂C	-2.36 (-2.19)	1.18 (1.09)
Cr₂C	-1.84 (-1.66)	0.91 (0.83)
Mo₂C	-1.61 (-1.59)	0.80 (0.79)
W₂C	-1.57 (-1.43)	0.79 (0.72)

Table S2. Energy of the FM solutions relative to the NM (or closed-shell) one (ΔE_{FM-NM} in meV) of $p(1\times 1)$ unit cell of M_2C ($M=Ti, V, Cr, Zr, Nb, Mo, Hf, Ta, \text{ and } W$) obtained from PBE, PBE0, and HSE06 functionals at the PBE optimized spin-polarized structures, as obtained with VASP and FHI-AIMS codes, the latter marked in italics. The negative sign indicates that the FM solution is more stable than the NM one. For diamagnetic MXenes the most stable FM solution is obtained by fixing the total number of unpaired electrons as described in the text.

MXene	PBE	PBE0	HSE06
Ti₂C^a	-135	-488	-420
	<i>-155</i>	<i>-506</i>	<i>-417</i>
Zr₂C	-36	-339	-274
	<i>20</i>	<i>-281</i>	<i>-197</i>
Hf₂C	60	-191	-143
	<i>79</i>	<i>-189</i>	<i>-105</i>
V₂C^b	769	151	128
	<i>797</i>	<i>189</i>	<i>222</i>
Nb₂C^b	1462	1246	1220
	<i>1497</i>	<i>1286</i>	<i>1301</i>
Ta₂C^b	2140	2134	2061
	<i>2187</i>	<i>2174</i>	<i>2177</i>
Cr₂C	-25	-1389	-1366
	<i>-15</i>	<i>-1238</i>	<i>-1122</i>
Mo₂C^c	349	292	295
	<i>395</i>	<i>324</i>	<i>330</i>
W₂C^c	812	883	841
	<i>867</i>	<i>1013</i>	<i>956</i>

^aThe values of Ti₂C from Ref. 45 of the main text have been also included to draw a clear and a wide picture of the influence of the metal atoms in these systems.

^bSpin-polarized solution fixing two unpaired electrons per unit cell.

^cSpin-polarized solution fixing one unpaired electron per unit cell.

Table S3. Energy of several FM solutions relative to the NM one (ΔE_{FM-NM} in meV, respectively) of $p(1\times 1)$ unit cell of paramagnetic Cr_2C and diamagnetic M_2C (M= Mo, and W) obtained from PBE, PBE0, and HSE06 functionals, at the PBE optimized spin-polarized structures for the corresponding GS, as obtained with VASP and FHI-AIMS codes, the latter marked in italics. These solutions have been estimated fixing the total net spin density per unit cell consistent with the Bader charges.

MXene	ΔE_{FM-NM}		
	PBE	PBE0	HSE06
Cr_2C^a	2236	-1320	-1268
	<i>2248</i>	<i>-1218</i>	<i>-1008</i>
Cr_2C^b	875	-1182	-1190
	<i>907</i>	<i>-1098</i>	<i>-1011</i>
Cr_2C^c	72	-77	-67
	<i>82</i>	<i>-45</i>	<i>-34</i>
Mo_2C^a	6644	4806	4886
	<i>6767</i>	<i>4967</i>	<i>5171</i>
Mo_2C^b	3222	2194	2224
	<i>3277</i>	<i>2222</i>	<i>2350</i>
W_2C^a	9118	7904	7875
	<i>9270</i>	<i>8190</i>	<i>8395</i>
W_2C^b	4534	3882	3839
	<i>4639</i>	<i>4051</i>	<i>4150</i>

^aSpin polarized solution fixing a value of 4 unpaired electrons per unit cell.

^bSpin polarized solution fixing a value of 3 unpaired electrons per unit cell.

^cSpin polarized solution fixing a value of 1 unpaired electron per unit cell.

Table S4. Energy of AFM solutions relative to the NM one (ΔE_{AFM-NM} in meV) of $p(1\times 1)$ unit cell of paramagnetic M_2C ($M=Ti, Zr, Hf,$ and Cr) obtained from PBE, PBE0, and HSE06 functionals at the PBE optimized spin-polarized structures with (a) VASP and (b) FHI-AIMS codes. The negative sign indicates that the AFM solutions are more stable than the NM one.

MXene	PBE	PBE0	HSE06
Ti₂C^a	-21	-99	-73
	-28	-103	-73
Zr₂C	-39	-90	-69
	-20	-128	-83
Hf₂C	-63	-81	-60
	-79	-89	-58
Cr₂C	45	-307	-263
	15	-425	-342

^aThe values of Ti₂C from Ref. 45 of the main text have been also included to draw a clear and a wide picture of the influence of the metal atoms in these systems.

Fig. S1. (a) AFM, (b) FM, and (c) NM projected DOS (in states/eV) for Ti_2C as obtained from calculations with the HSE06 functional at the PBE optimized $p(1\times 1)$ unit cell. The energy is referenced to the Fermi level (in eV).

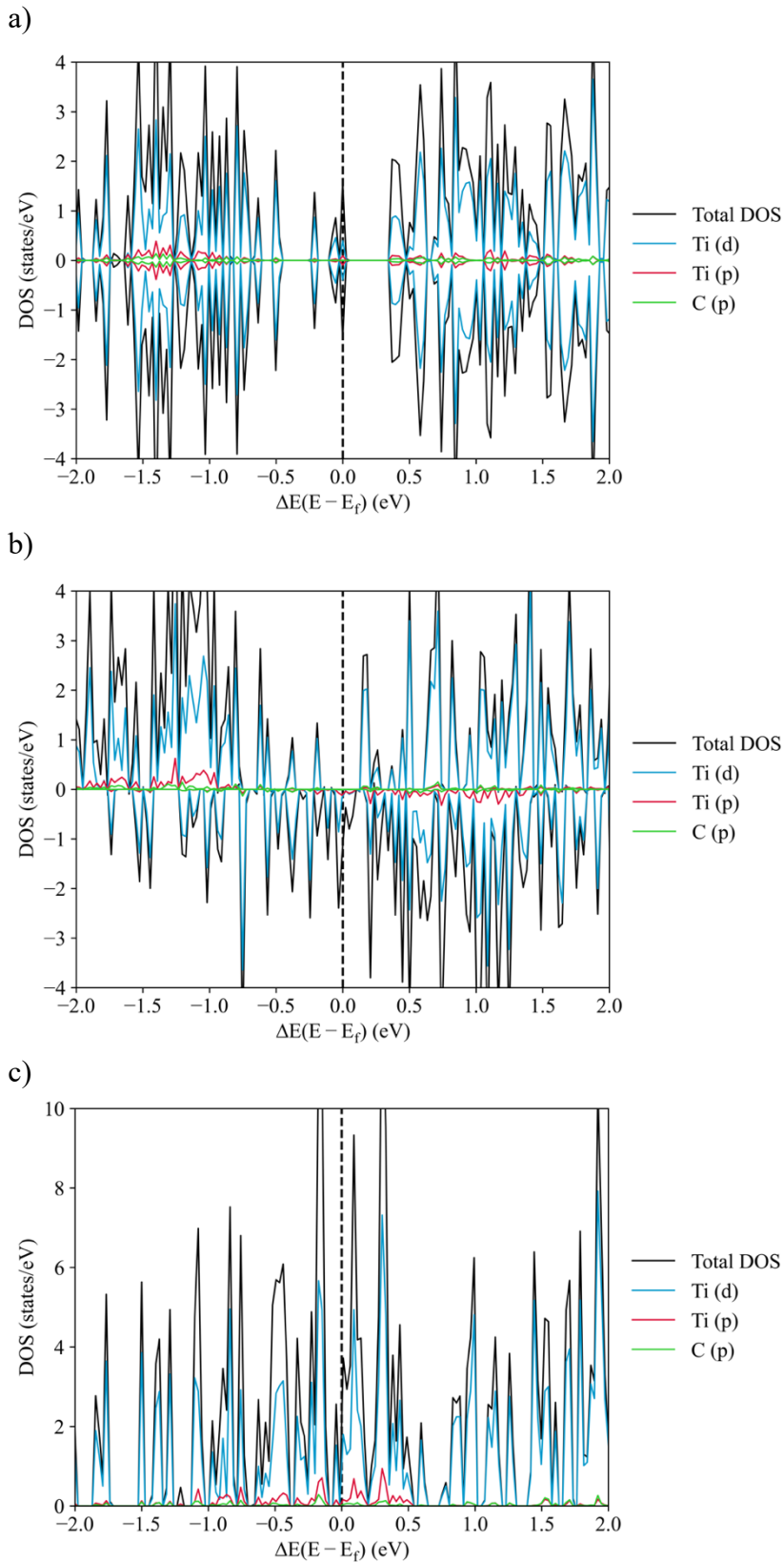


Fig. S2. (a) FM and (b) NM projected DOS (in states/eV) for V₂C as obtained from calculations with the HSE06 functional at the PBE optimized $p(1\times 1)$ unit cell. The energy is referenced to the Fermi level (in eV).

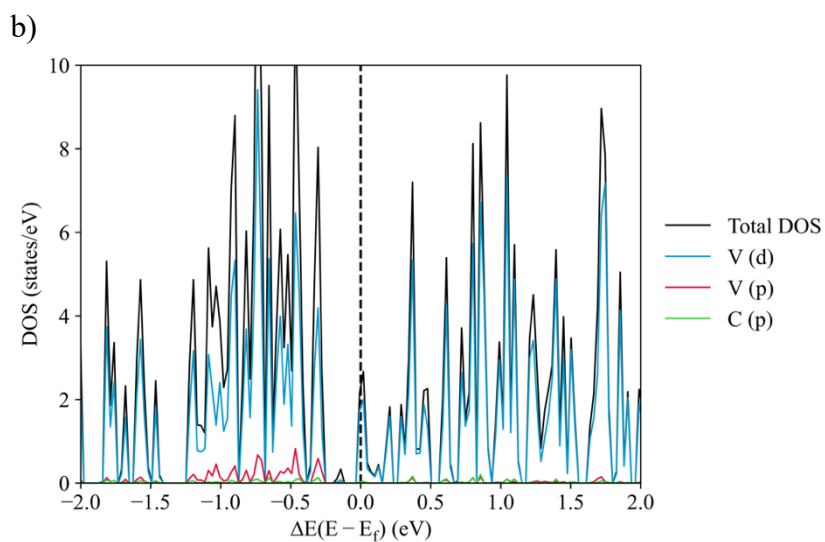
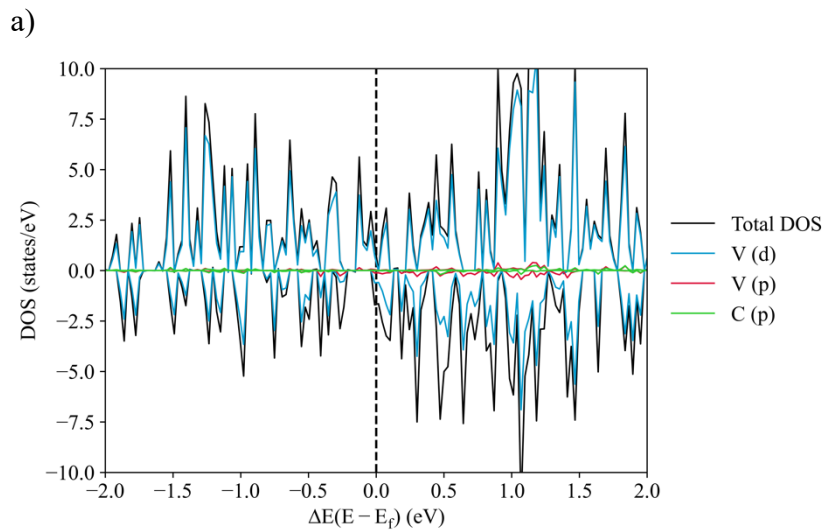


Fig. S3. (a) AFM, (b) FM, and (c) NM projected DOS (in states/eV) for Cr₂C as obtained from calculations with the HSE06 functional at the PBE optimized $p(1\times 1)$ unit cell. The energy is referenced to the Fermi level (in eV).

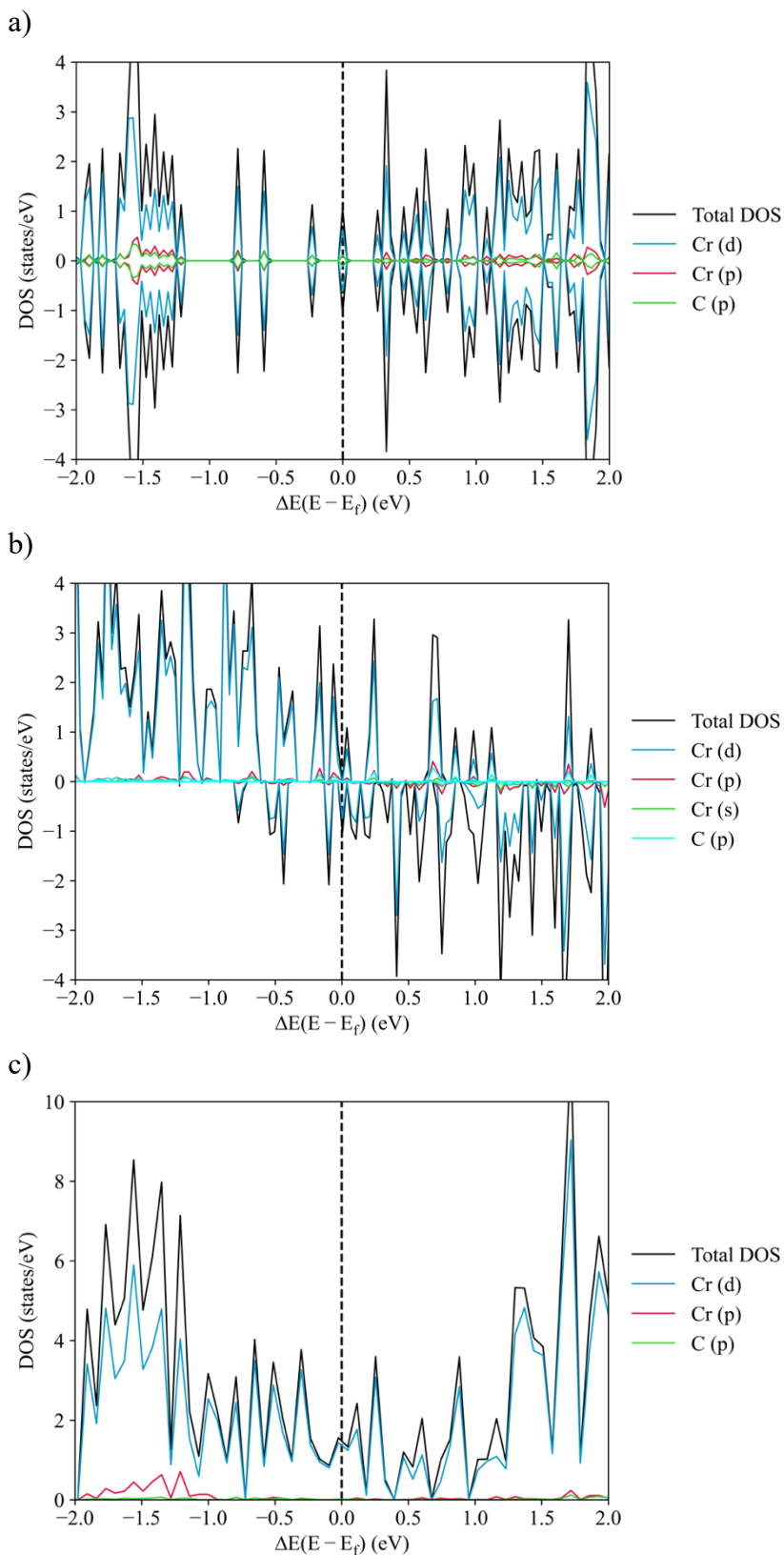


Fig. S4. (a) AFM, (b) FM, and (c) NM projected DOS (in states/eV) for Zr_2C as obtained from calculations with the HSE06 functional at the PBE optimized $p(1\times 1)$ unit cell. The energy is referenced to the Fermi level (in eV).

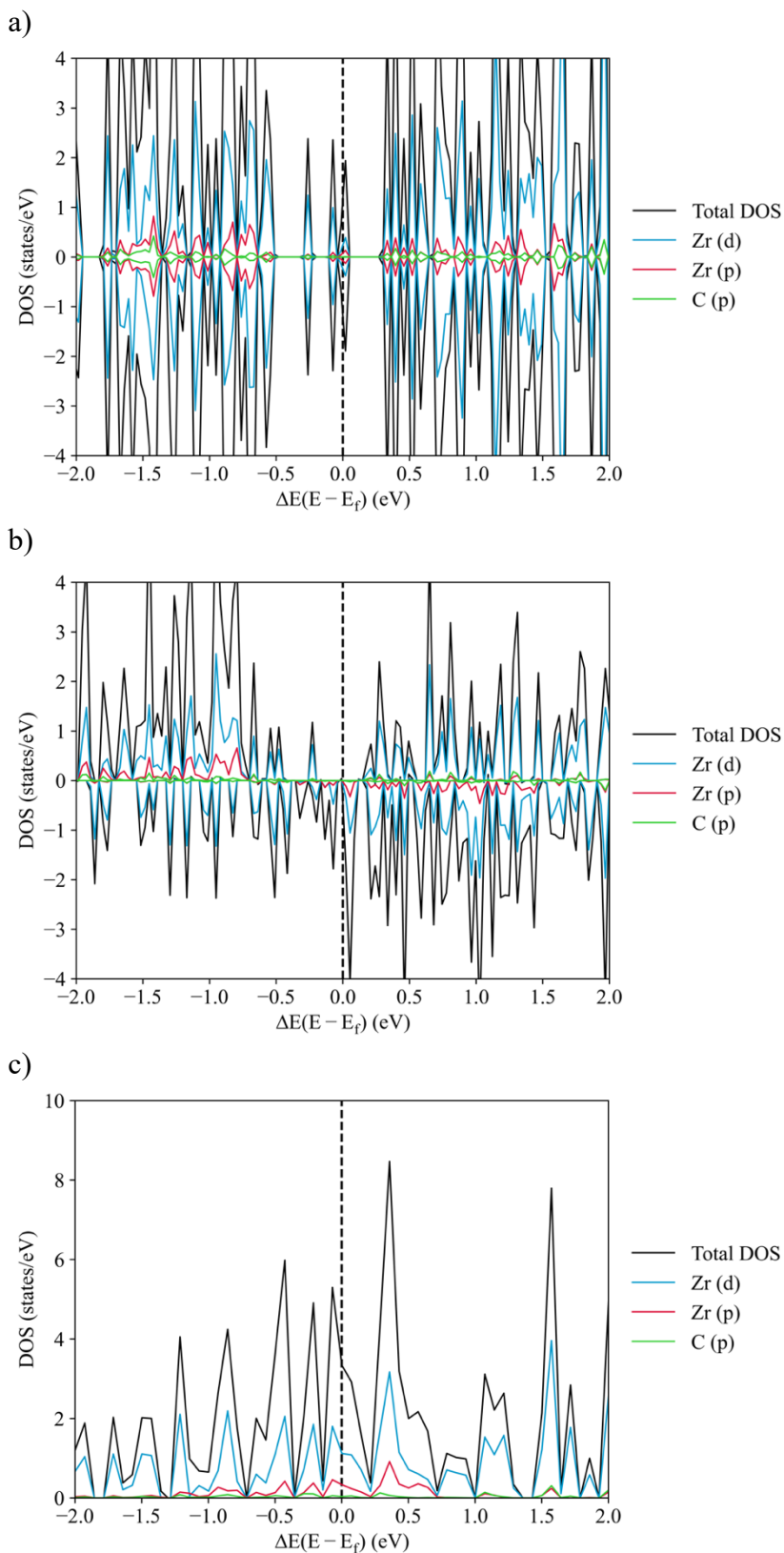


Fig. S5. (a) FM and (b) NM projected DOS (in states/eV) for Nb₂C as obtained from calculations with the HSE06 functional at the PBE optimized $p(1\times 1)$ unit cell. The energy is referenced to the Fermi level (in eV).

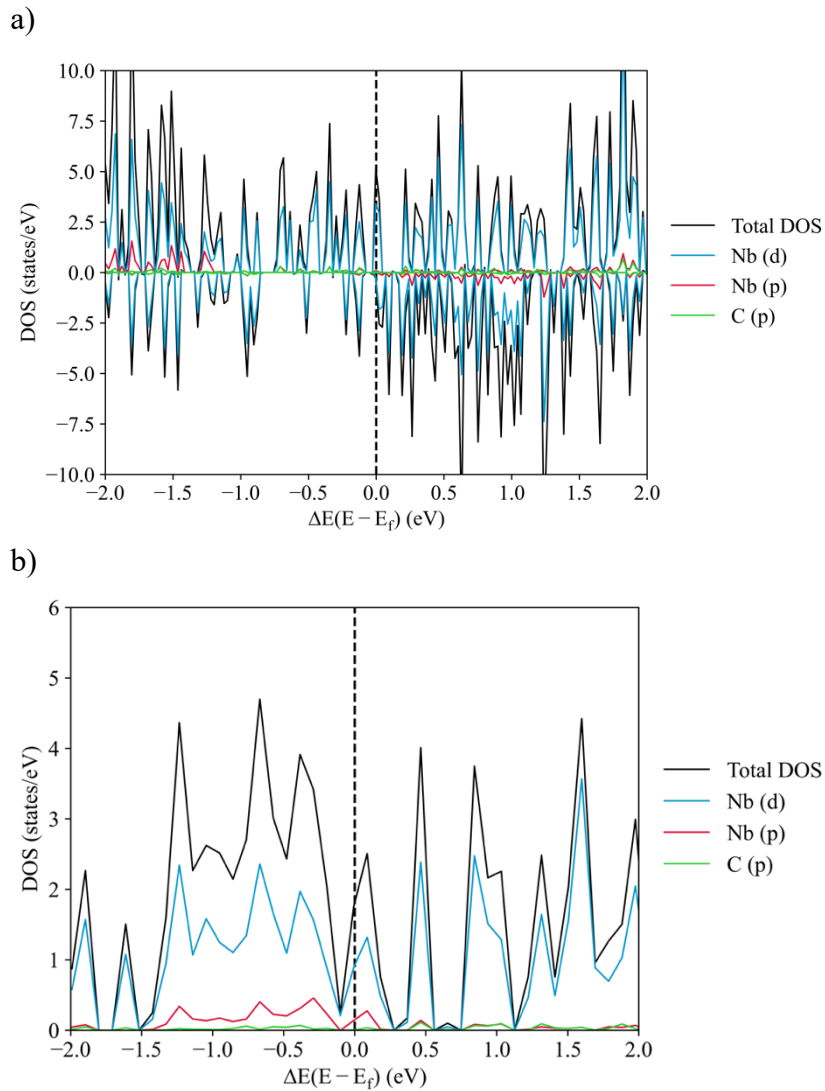
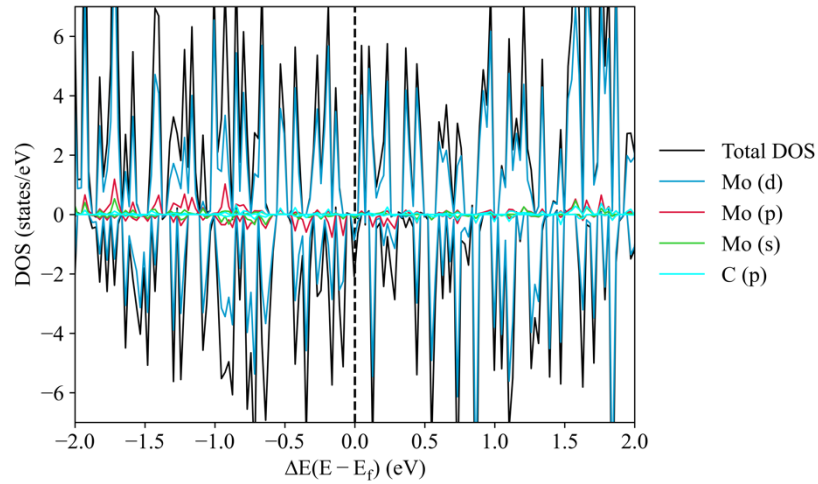


Fig. S6. (a) FM and (b) NM projected DOS (in states/eV) for Mo₂C as obtained from calculations with the HSE06 functional at the PBE optimized $p(1\times 1)$ unit cell. The energy is referenced to the Fermi level (in eV).

a)



b)

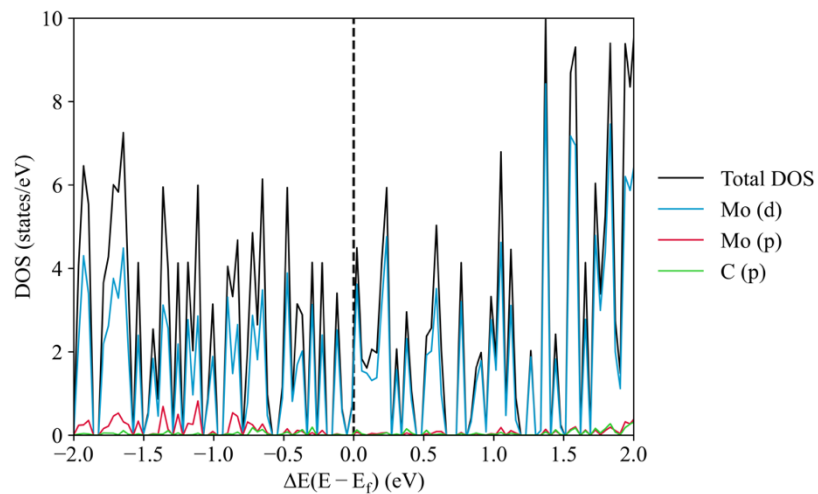


Fig. S7. (a) AFM, (b) FM, and (c) NM projected DOS (in states/eV) for Hf₂C as obtained from calculations with the HSE06 functional at the PBE optimized $p(1\times 1)$ unit cell. The energy is referenced to the Fermi level (in eV).

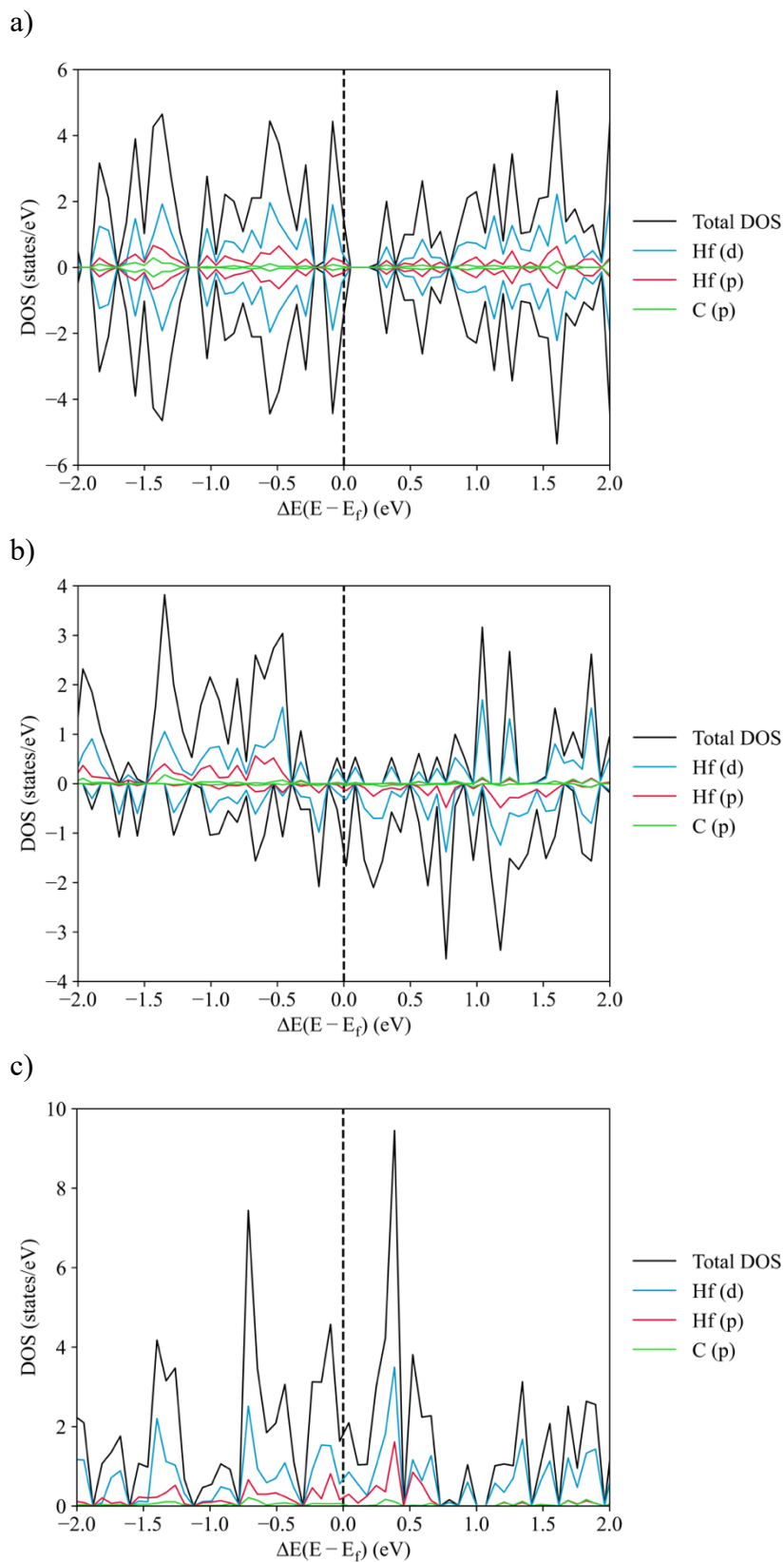
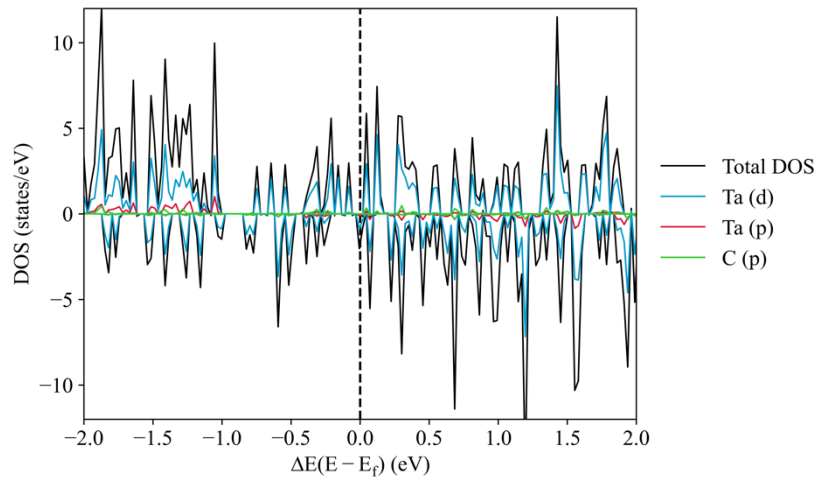


Fig. S8. (a) FM and (b) NM projected DOS (in states/eV) for Ta₂C as obtained from calculations with the HSE06 functional at the PBE optimized $p(1\times 1)$ unit cell. The energy is referenced to the Fermi level (in eV).

a)



b)

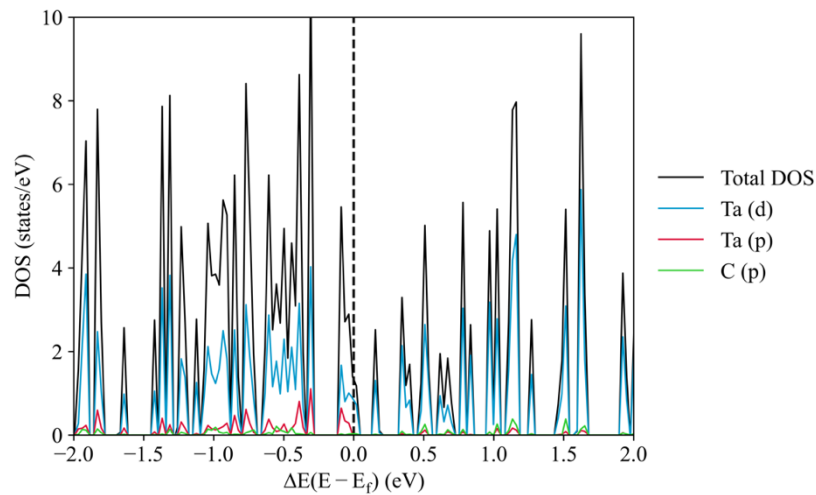
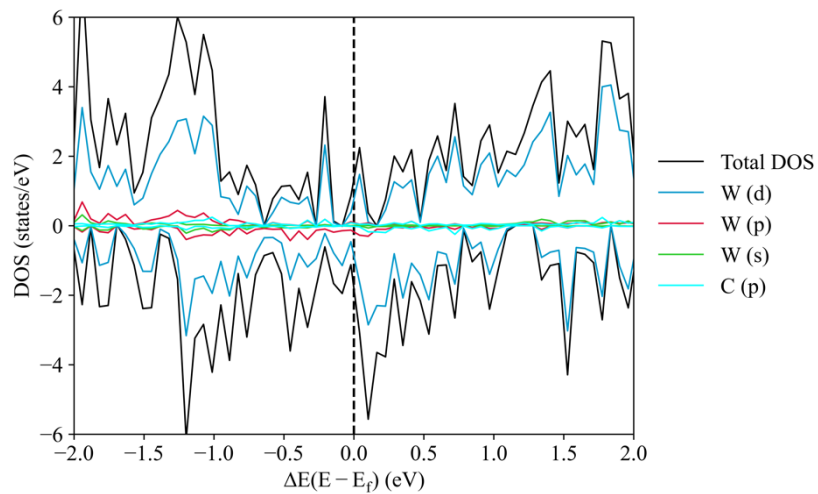


Fig. S9. (a) FM and (b) NM projected DOS (in states/eV) for W_2C as obtained from calculations with the HSE06 functional at the PBE optimized $p(1 \times 1)$ unit cell. The energy is referenced to the Fermi level (in eV).

a)



b)

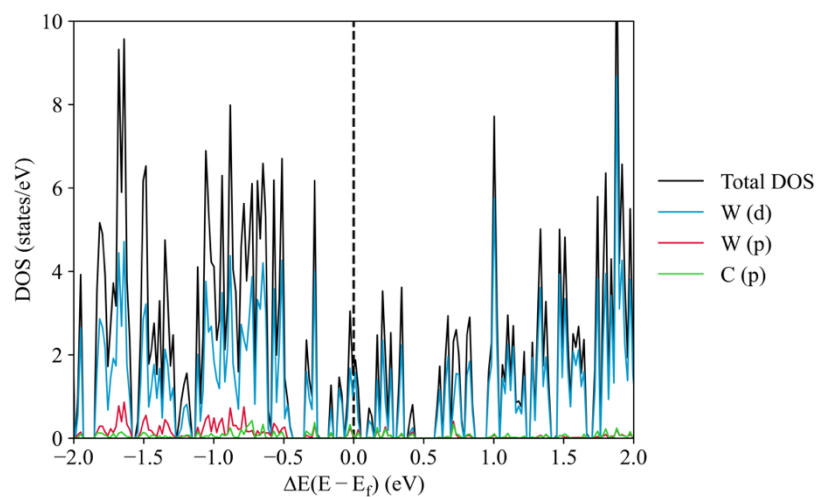


Fig. S10. Phonon dispersion of diamagnetic (left) and paramagnetic (right) solutions of Ti₂C as predicted from the calculations with the PBE functional.

