Electronic Supplementary Material (ESI) for Nanoscale Horizons.

Computational design of double transition metal MXenes with intrinsic magnetic properties

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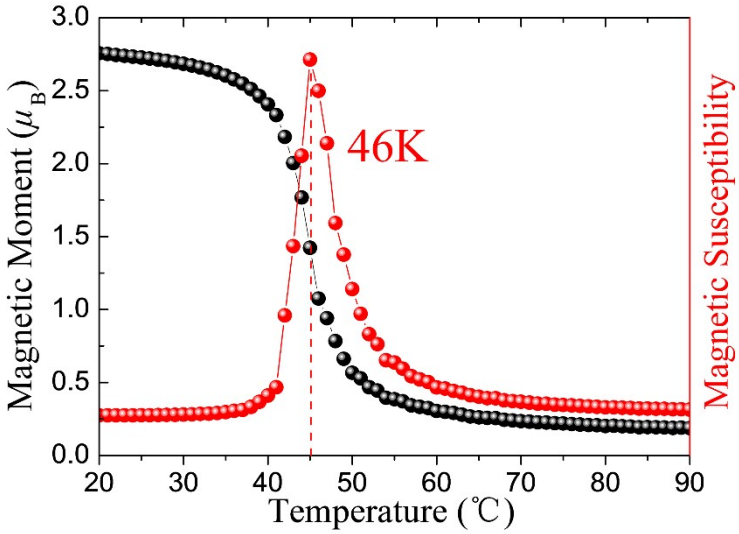

Figure S1. Magnetic moment of Cr atoms and magnetic susceptibility as a function of temperature for CrI_3 monolayers.

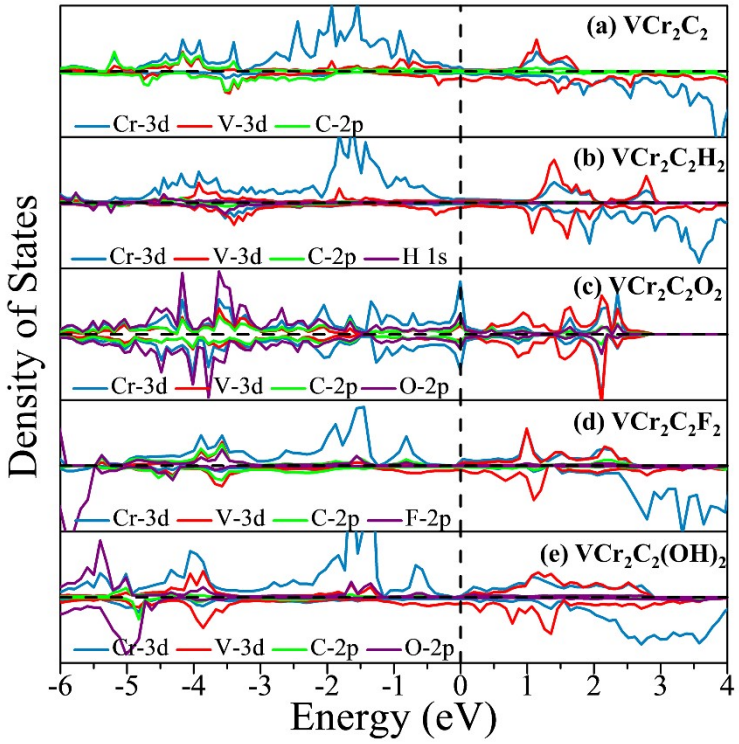

Figure S2. The spin-revolved density of states (DOS) of $VCr_2C_2T_x$ (T = H, O, F, OH, or bare) MXenes.

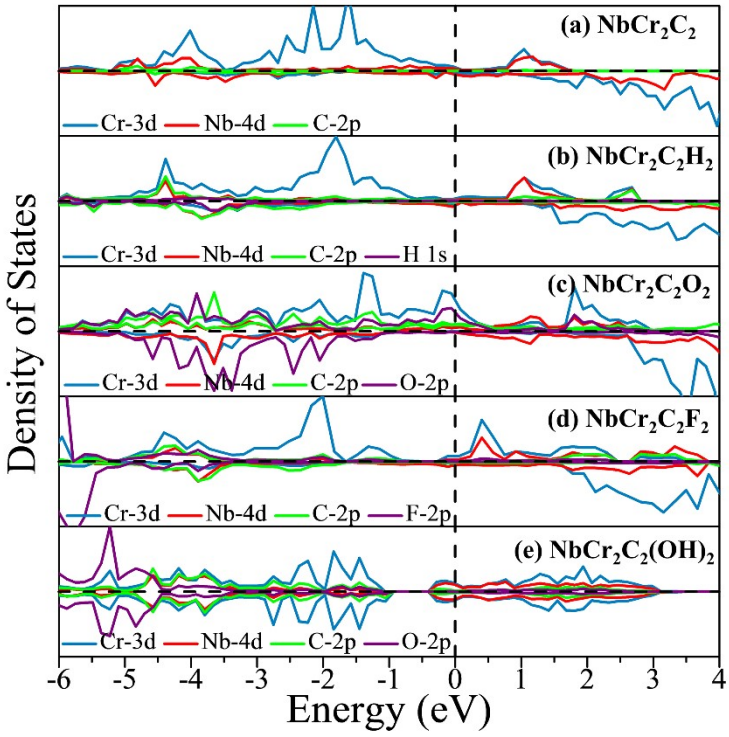

Figure S3. The spin-revolved density of states (DOS) of $NbCr_2C_2T_x$ (T = H, O, F, OH, or bare) MXenes.

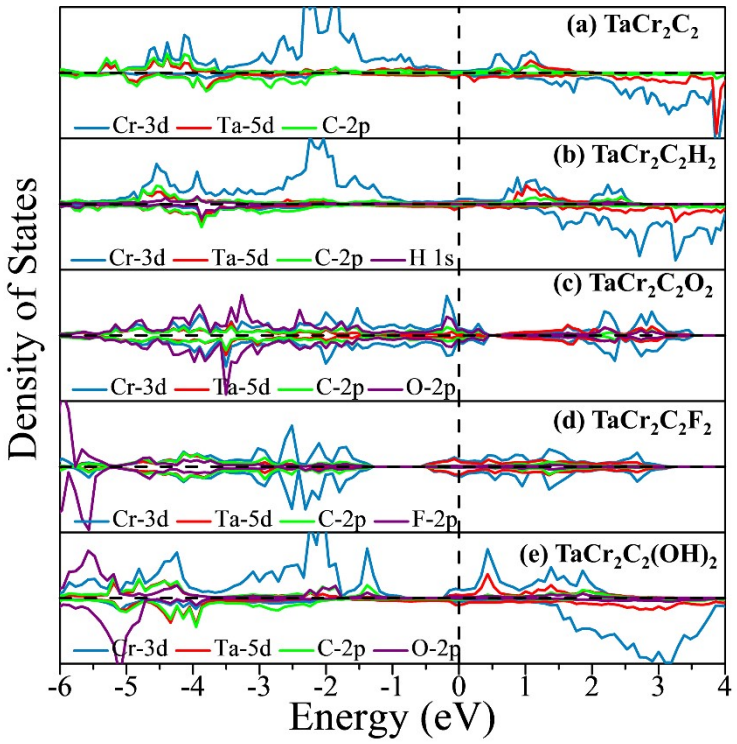

Figure S4. The spin-revolved density of states (DOS) of $TaCr_2C_2T_x$ (T = H, O, F, OH, or bare) MXenes.

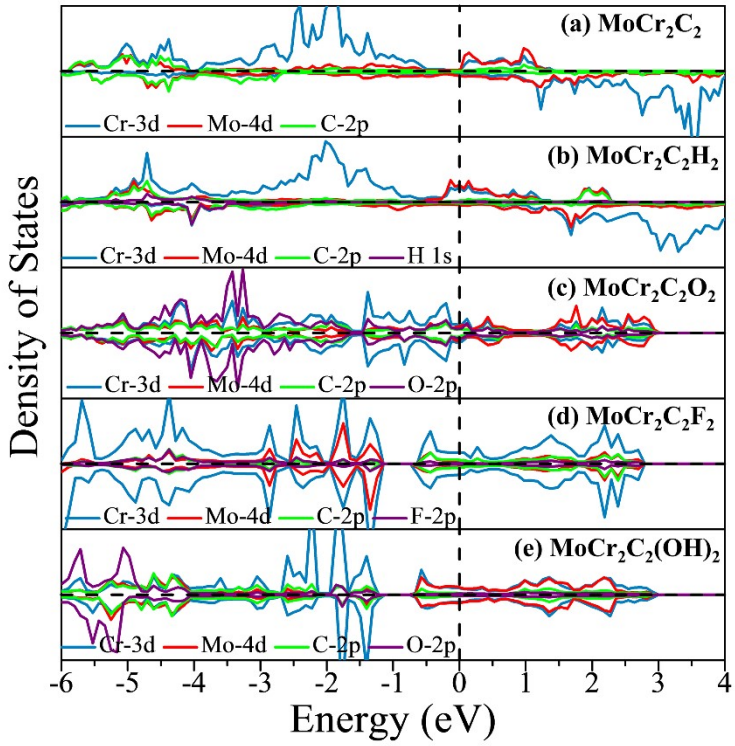

Figure S5. The spin-revolved density of states (DOS) of $MoCr_2C_2T_x$ (T = H, O, F, OH, or bare) MXenes.

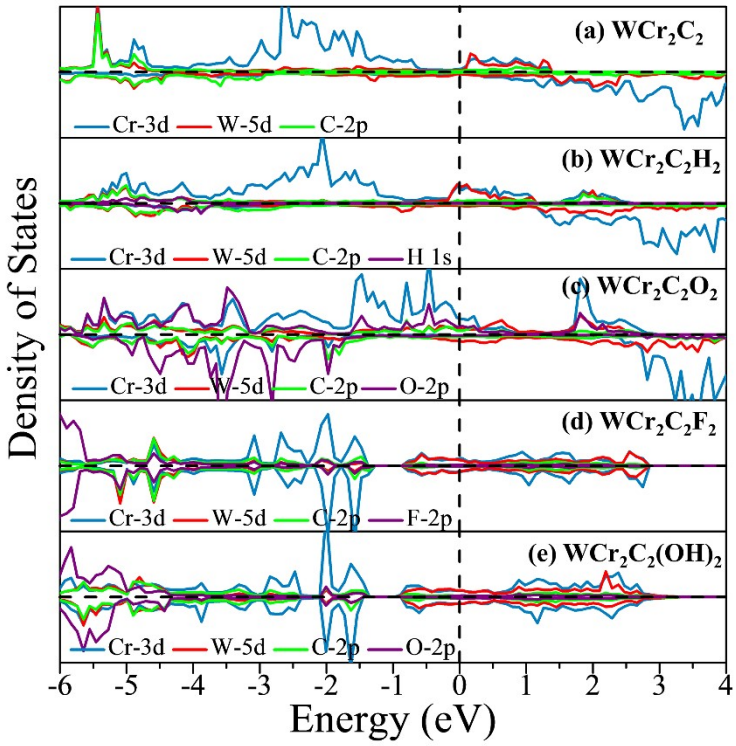

Figure S6. The spin-revolved density of states (DOS) of $WCr_2C_2T_x$ (T = H, O, F, OH, or bare) MXenes.

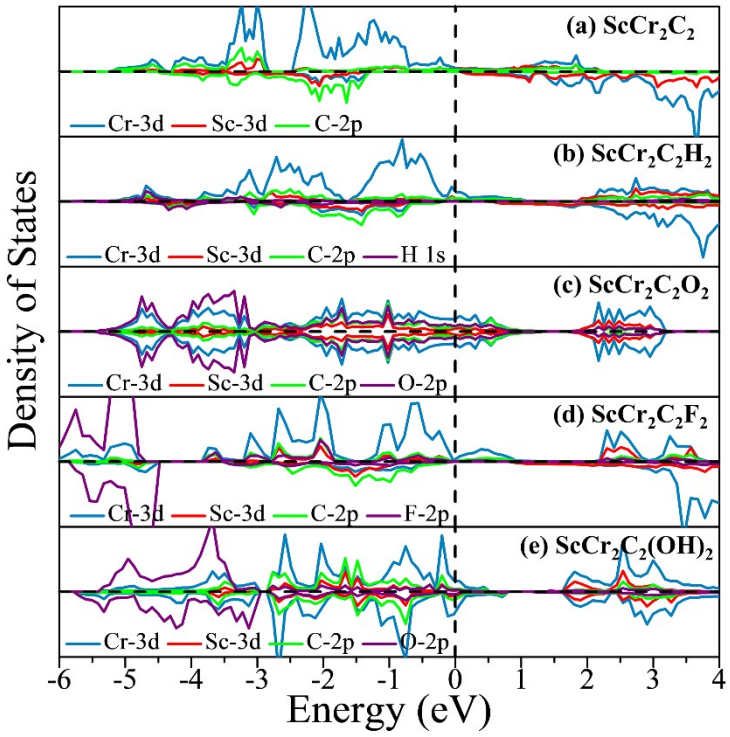

Figure S7. The spin-revolved density of states (DOS) of $ScCr_2C_2T_x$ (T = H, O, F, OH, or bare) MXenes.

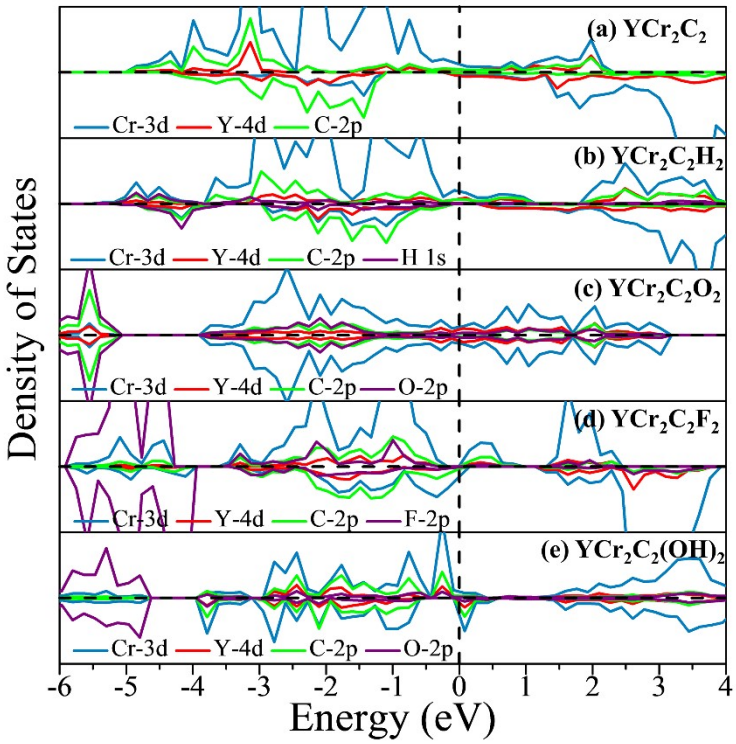

Figure S8. The spin-revolved density of states (DOS) of $YCr_2C_2T_x$ (T = H, O, F, OH, or bare) MXenes.

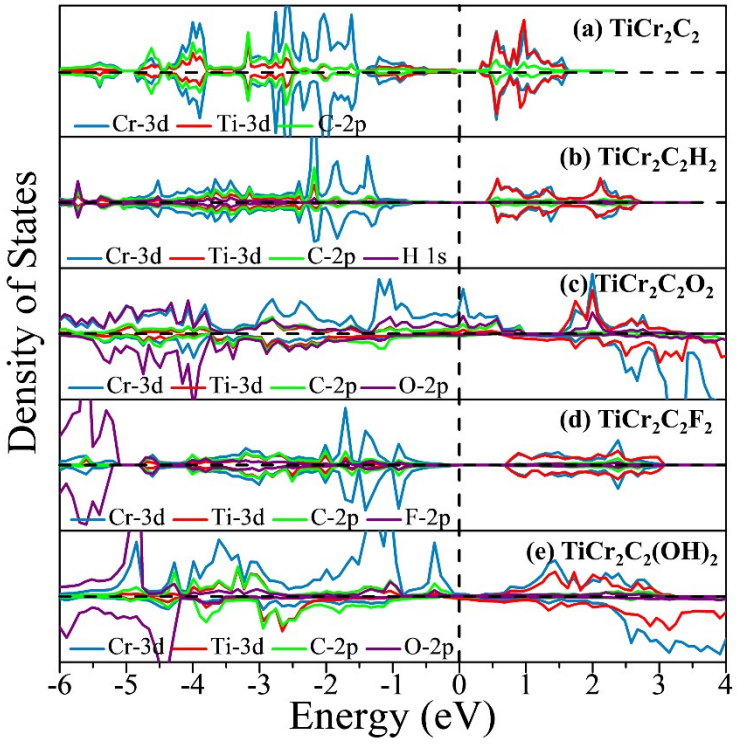

Figure S9. The spin-revolved density of states (DOS) of $TiCr_2C_2T_x$ (T = H, O, F, OH, or bare) MXenes.

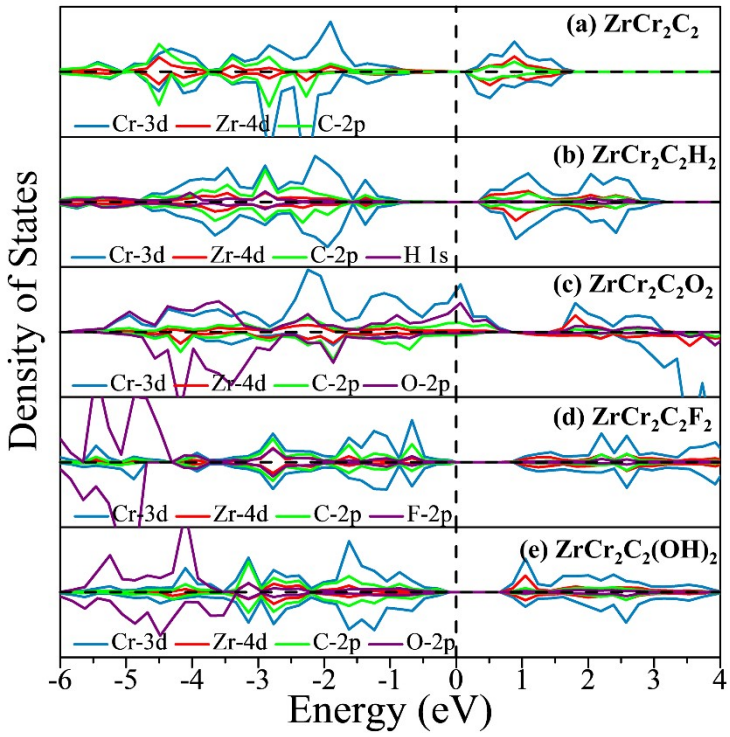

Figure S10. The spin-revolved density of states (DOS) of $ZrCr_2C_2T_x$ (T = H, O, F, OH, or bare) MXenes.

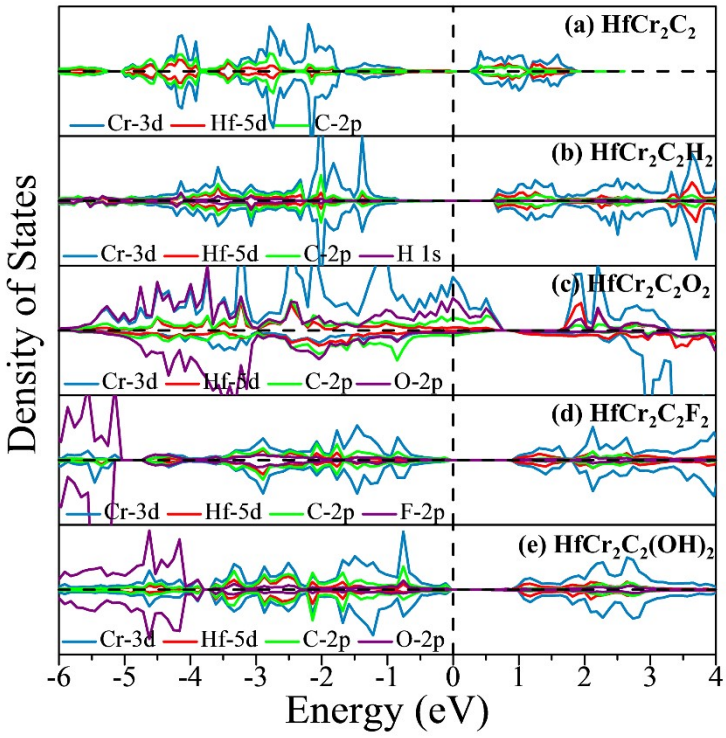

Figure S11. The spin-revolved density of states (DOS) of $HfCr_2C_2T_x$ (T = H, O, F, OH, or bare) MXenes.

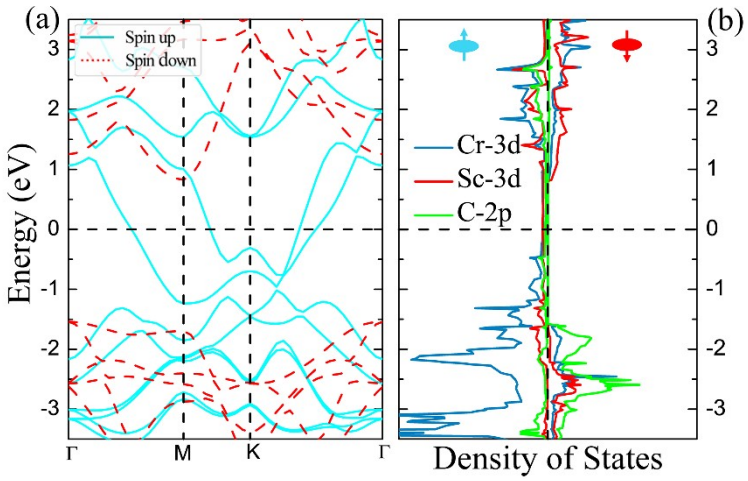

Figure S12. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $ScCr_2C_2$ obtained by HSE06 approach.

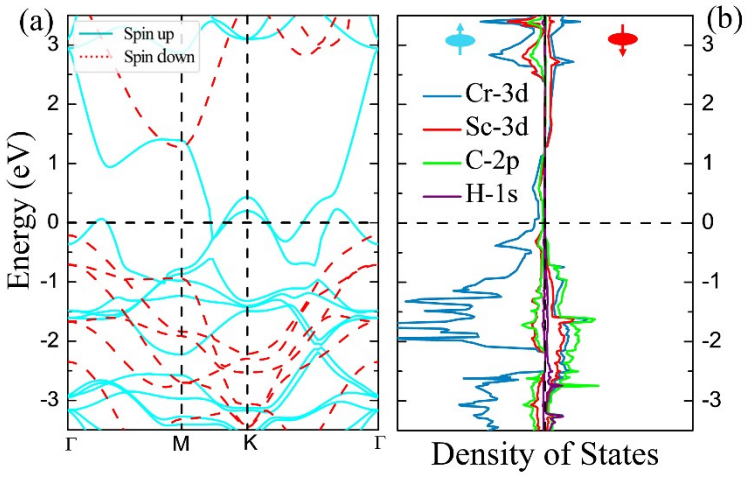

Figure S13. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $ScCr_2C_2H_2$ obtained by HSE06 approach.

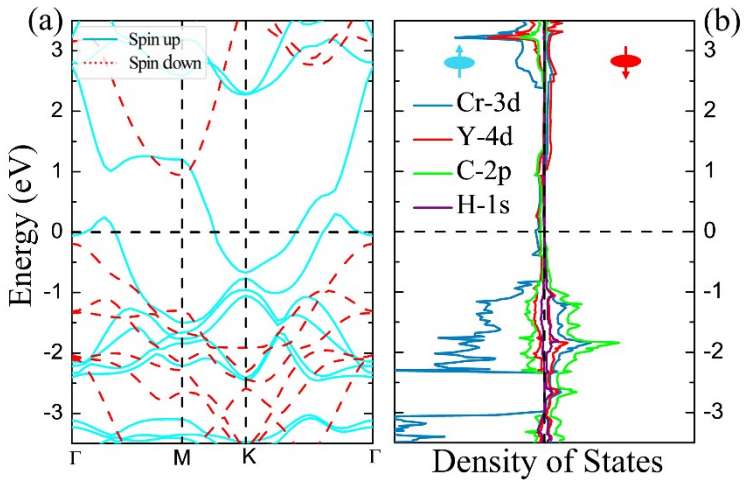

Figure S14. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $YCr_2C_2H_2$ obtained by HSE06 approach.

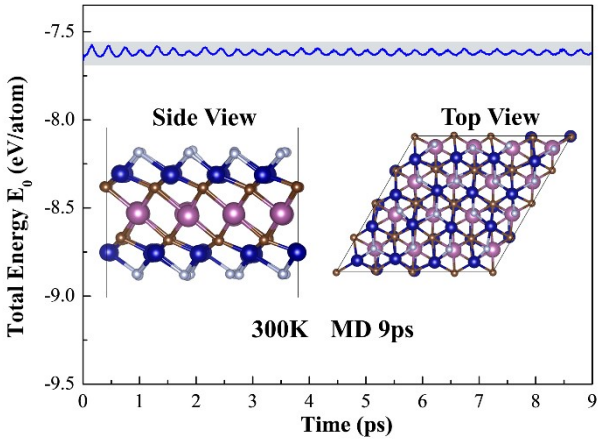

Figure S15. Total energy changes of $ScCr_2C_2F_2$ by AIMD calculations from 0 to 9 ps at 300K, the insets are the structure snapshots of $ScCr_2C_2F_2$ at 9 ps.

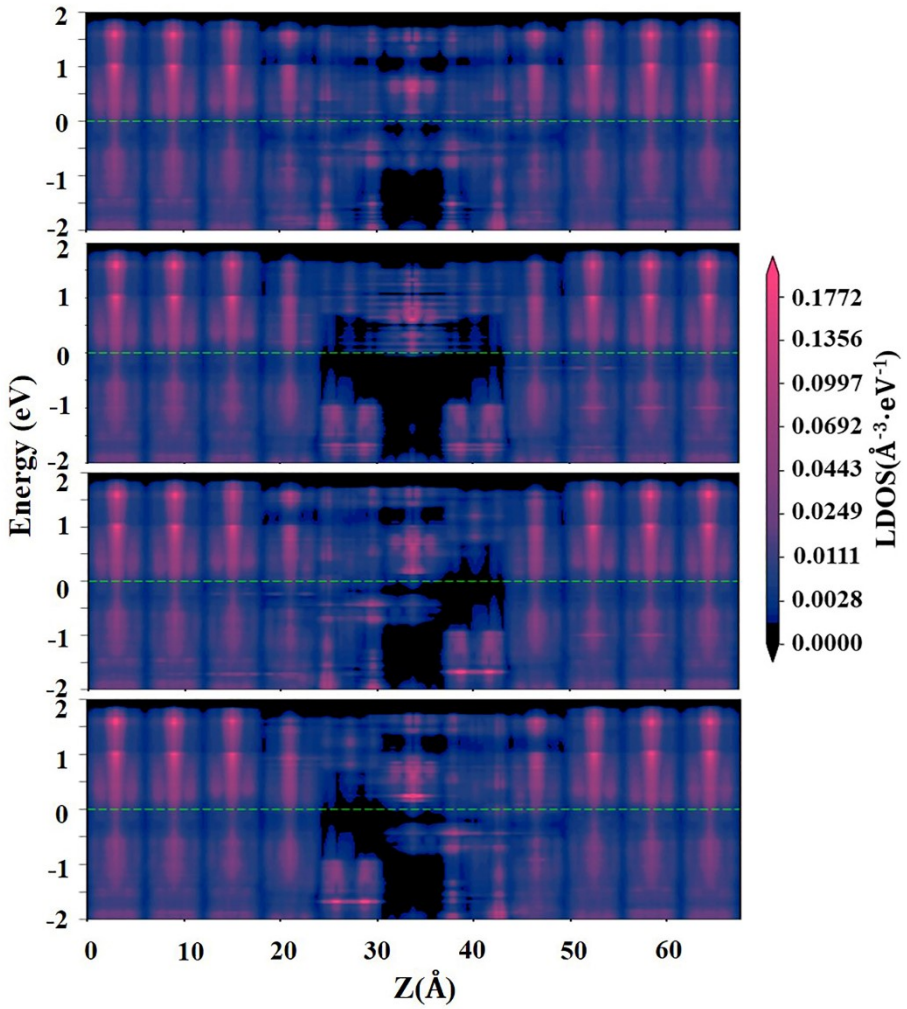

Figure S16. The Spin-resolved projected local density of states (PLDOS) of $ScCr_2C_2$/2H-MoS_2/$ScCr_2C_2$ magnetic tunnel junction (MTJ) along transport direction, (a) and (b) stand for the spin-up and spin-down electrons in parallel configuration (PC), (c) and (d) represent the spin-up and spin-down electrons in antiparallel configuration (APC). The Fermi level is labeled with green dashed line.

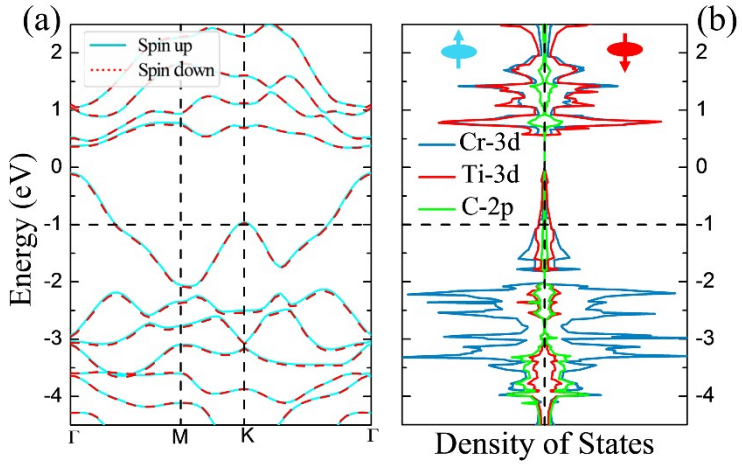

Figure S17. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $TiCr_2C_2$ obtained by HSE06 approach.

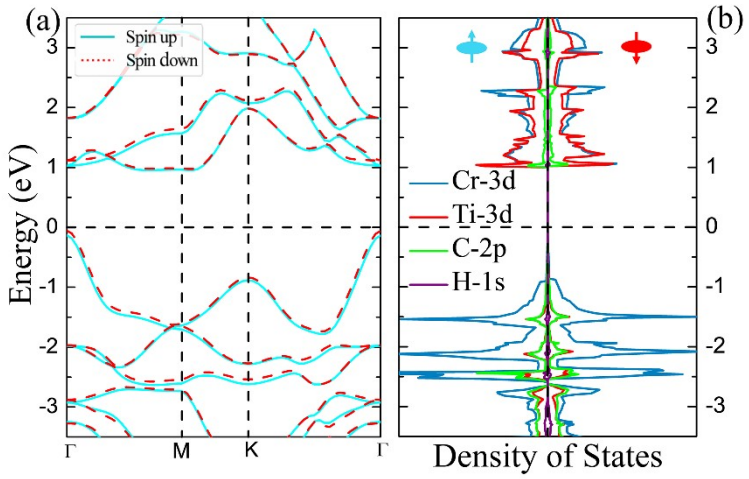

Figure S18. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $TiCr_2C_2H_2$ obtained by HSE06 approach.

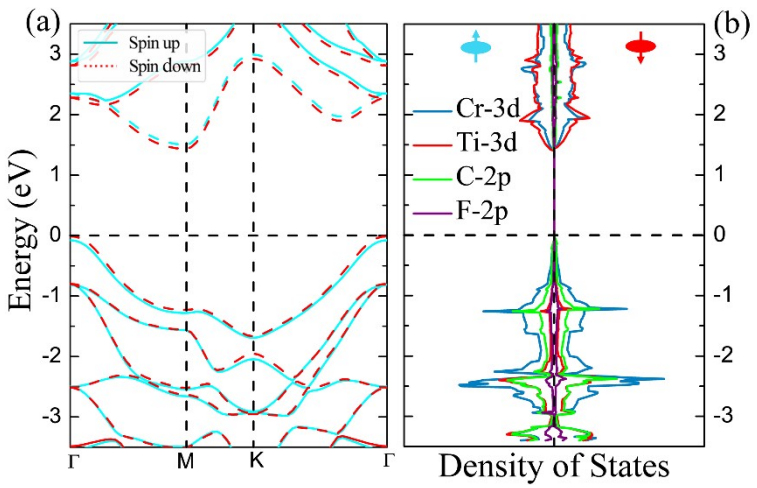

Figure S19. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $TiCr_2C_2F_2$ obtained by HSE06 approach.

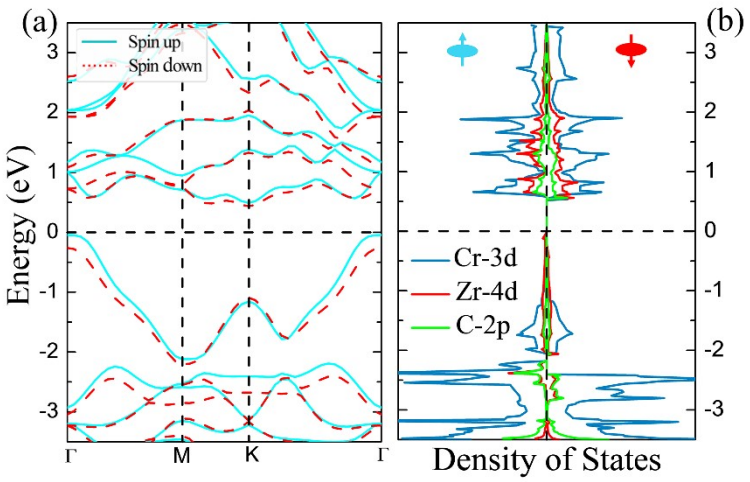

Figure S20. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $ZrCr_2C_2$ obtained by HSE06 approach.

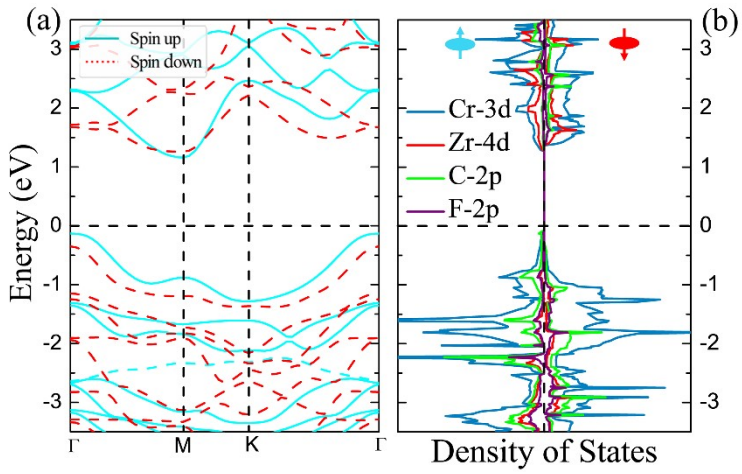

Figure S21. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $ZrCr_2C_2F_2$ obtained by HSE06 approach.

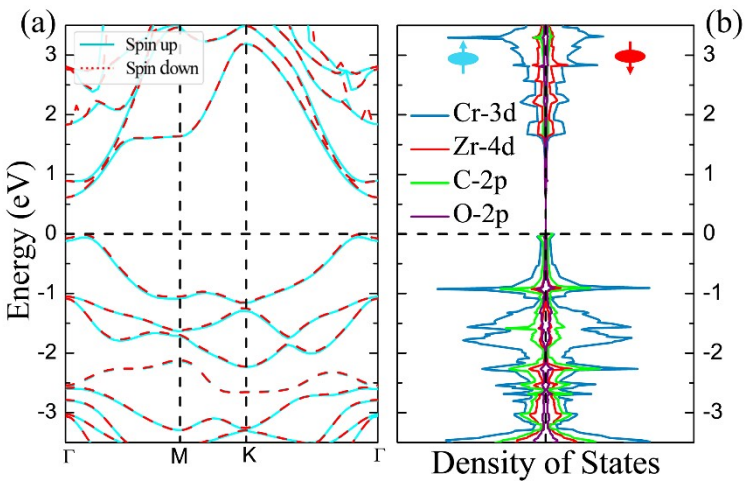

Figure S22. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $ZrCr_2C_2(OH)_2$ obtained by HSE06 approach.

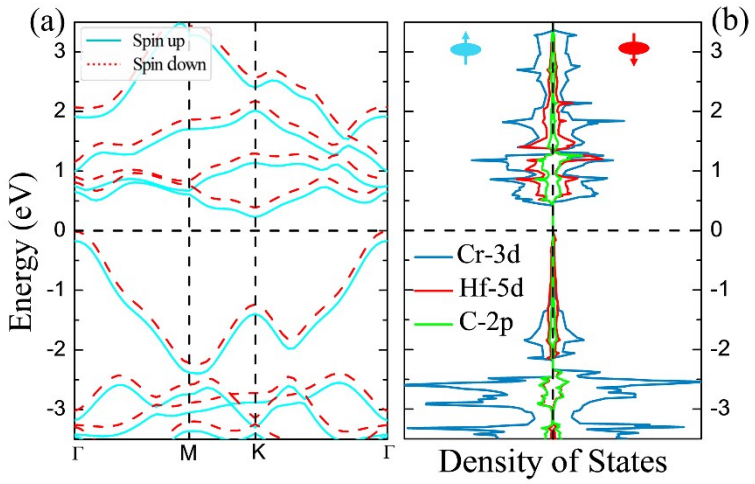

Figure S23. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $HfCr_2C_2$ obtained by HSE06 approach.

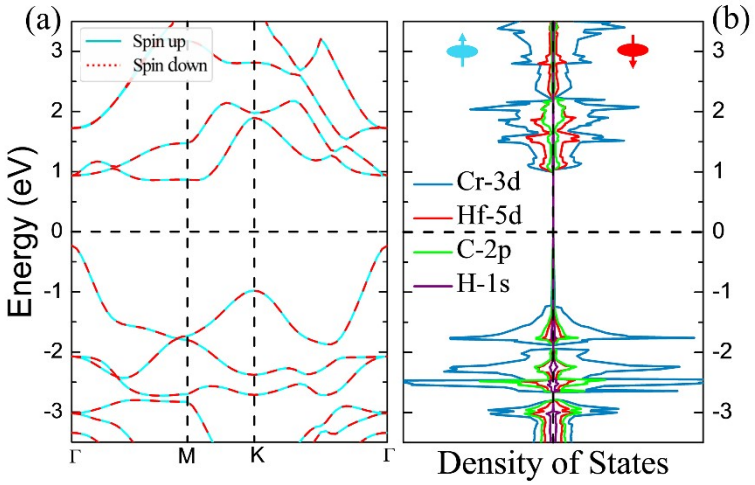

Figure S24. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $HfCr_2C_2H_2$ obtained by HSE06 approach.

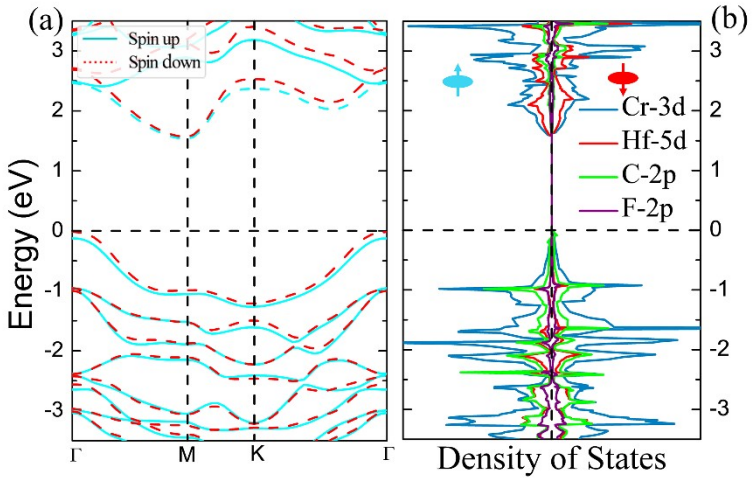

Figure S25. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $HfCr_2C_2F_2$ obtained by HSE06 approach.

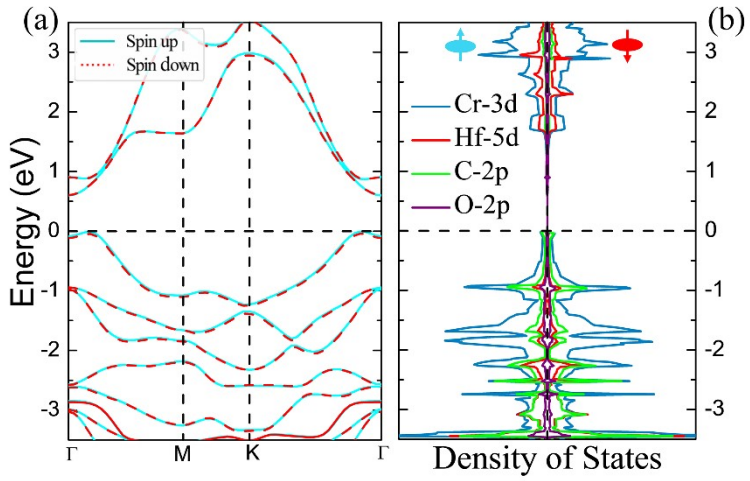

Figure S26. (a) Spin-dependent electronic band structure and (b) spin-resolved density of states of $HfCr_2C_2(OH)_2$ obtained by HSE06 approach.

Table S1. The calculated energies (eV) for the most stable model and preferred magnetic orders of $ScCr_2C_2T_x$ (T = H, O, F, OH, or bare). The most stable model is highlighted in bold and the preferred magnetic orders are marked with yellow background.

$ScCr_2C_2$		Model I	Model II	Model III
Bare	NM	-70.572		
	AFM1	-77.019		
	AFM2	-76.646		
	AFM3	-76.488		
	FM	**-77.325**		
H	NM	-84.758	**-86.205**	-85.456
	AFM1	-88.899	**-91.370**	-90.183
	AFM2	-87.998	**-90.918**	-89.617
	AFM3	-87.998	**-90.831**	-89.496
	FM	-89.009	**-91.481**	-90.233
O	NM	**-102.193**	-101.348	-101.968
	AFM1	-103.532	-103.822	**-104.768**
	AFM2	-102.905	**-105.060**	-104.319
	AFM3	-102.887	**-105.531**	-104.292
	FM	-103.522	**-103.787**	-103.855
F	NM	-92.446	**-92.602**	-92.142
	AFM1	-97.181	**-99.608**	-98.430
	AFM2	-96.846	**-99.675**	-98.134
	AFM3	-96.538	**-99.490**	-98.195
	FM	-96.687	**-100.046**	-98.460
OH	NM	**-116.951**	-115.113	-115.799
	AFM1	-121.313	**-123.734**	-122.223
	AFM2	-119.817	**-123.142**	-121.097
	AFM3	-120.112	**-123.360**	-118.902
	FM	-121.304	**-123.360**	-122.545

Table S2. The calculated energies (eV) for the most stable model and preferred magnetic orders of $YCr_2C_2T_x$ (T = H, O, F, OH, or bare).

YCr_2C_2		Model I	Model II	Model III
Bare	NM	-67.726		
	AFM1	-76.424		
	AFM2	-75.981		
	AFM3	-75.885		
	FM	**-76.542**		
H	NM	-82.189	**-83.920**	-82.991
	AFM1	-89.548	**-89.640**	-89.276
	AFM2	-86.200	**-89.206**	-88.744
	AFM3	-86.224	**-89.201**	-88.792
	FM	-87.256	**-89.772**	-88.470
O	NM	**-99.499**	-99.431	-99.135
	AFM1	-101.959	**-103.892**	-101.213
	AFM2	**-104.105**	-103.443	-104.098
	AFM3	**-104.419**	-103.312	-103.956
	FM	-102.528	-104.062	**-104.210**
F	NM	-90.438	**-90.753**	-90.452
	AFM1	-98.409	**-98.600**	-97.079
	AFM2	-94.946	**-98.268**	-96.408
	AFM3	-95.144	**-98.454**	-96.804
	FM	-95.661	**-98.538**	-97.047
OH	NM	-112.797	**-115.325**	-114.516
	AFM1	-116.883	**-121.816**	-120.281
	AFM2	-118.049	**-121.443**	-120.126
	AFM3	-118.402	**-121.075**	-119.929
	FM	-117.893	**-121.279**	-120.414

Table S3. The calculated energies (eV) for the most stable model and preferred magnetic orders of $TiCr_2C_2T_x$ (T = H, O, F, OH, or bare).

$TiCr_2C_2$		Model I	Model II	Model III
Bare	NM	-69.002		
	AFM1	**-75.070**		
	AFM2	-74.535		
	AFM3	-74.644		
	FM	-74.785		
H	NM	-83.513	**-84.654**	-84.053
	AFM1	-88.451	**-90.929**	-89.669
	AFM2	-87.674	**-90.351**	-89.020
	AFM3	-87.495	**-90.308**	-88.895
	FM	-88.225	**-90.649**	-89.428
O	NM	**-101.235**	-100.065	-100.737
	AFM1	-102.246	**-105.110**	-103.976
	AFM2	-103.033	**-105.153**	-104.117
	AFM3	-102.670	**-105.234**	-103.679
	FM	-102.403	**-105.285**	-104.269
F	NM	**-90.670**	-90.112	-90.501
	AFM1	-96.553	**-99.550**	-97.949
	AFM2	-94.159	**-99.063**	-97.702
	AFM3	-94.370	**-98.771**	-97.427
	FM	-96.310	**-99.326**	-97.881
OH	NM	**-114.966**	-113.970	-114.683
	AFM1	-119.879	**-122.642**	-120.847
	AFM2	-119.574	**-121.563**	-118.133
	AFM3	-118.822	**-122.363**	-120.336
	FM	-117.645	**-122.754**	-121.512

Table S4. The calculated energies (eV) for the most stable model and preferred magnetic orders of $ZrCr_2C_2T_x$ (T = H, O, F, OH, or bare).

$ZrCr_2C_2$		Model I	Model II	Model III
Bare	NM	-75.323		
	AFM1	**-82.732**		
	AFM2	-82.184		
	AFM3	-81.180		
	FM	-82.416		
H	NM	-89.131	**-90.775**	-89.944
	AFM1	-95.046	-97.770	**-97.794**
	AFM2	-94.185	**-97.053**	-95.377
	AFM3	-93.936	**-96.918**	-95.406
	FM	-94.839	**-97.433**	-96.171
O	NM	-106.139	**-106.755**	-106.646
	AFM1	-109.098	**-111.830**	-110.507
	AFM2	-109.303	**-111.508**	-108.849
	AFM3	-109.272	**-111.302**	-110.307
	FM	-109.354	**-111.947**	-110.579
F	NM	-96.960	**-97.891**	-97.221
	AFM1	-103.022	**-106.888**	-105.023
	AFM2	-102.842	**-105.969**	-104.350
	AFM3	-100.769	**-105.953**	-104.554
	FM	-103.133	**-106.404**	-104.757
OH	NM	-121.412	**-121.425**	-120.248
	AFM1	-126.985	**-130.462**	-128.744
	AFM2	-127.012	**-129.251**	-127.476
	AFM3	-126.881	**-129.771**	-127.883
	FM	-126.937	**-129.927**	-128.549

Table S5. The calculated energies (eV) for the most stable model and preferred magnetic orders of $HfCr_2C_2T_x$ (T = H, O, F, OH, or bare).

$HfCr_2C_2$		Model I	Model II	Model III
Bare	NM	-78.418		
	AFM1	**-86.015**		
	AFM2	-85.419		
	AFM3	-85.174		
	FM	-85.517		
H	NM	-92.614	**-94.140**	-93.382
	AFM1	-101.089	**-101.216**	-99.851
	AFM2	-97.423	**-100.324**	-98.945
	AFM3	-97.690	**-100.544**	-99.111
	FM	-98.242	**-100.873**	-99.620
O	NM	-109.822	**-109.928**	-109.738
	AFM1	-112.320	**-115.381**	-114.062
	AFM2	-112.321	**-113.455**	-113.982
	AFM3	-111.628	**-114.960**	-113.845
	FM	-112.427	**-115.431**	-114.079
F	NM	-100.267	**-101.144**	-100.571
	AFM1	-106.875	**-110.408**	-108.428
	AFM2	-106.188	**-109.453**	-107.833
	AFM3	-106.557	**-109.503**	-107.868
	FM	-106.438	**-109.898**	-108.333
OH	NM	-123.896	-124.303	**-124.967**
	AFM1	-130.688	**-133.707**	-131.823
	AFM2	-130.568	**-132.878**	-130.053
	AFM3	-130.015	**-133.049**	-129.643
	FM	-130.563	**-133.216**	-131.554

Table S6. The calculated energies (eV) for the most stable model and preferred magnetic orders of $VCr_2C_2T_x$ (T = H, O, F, OH, or bare).

VCr_2C_2		Model I	Model II	Model III
Bare	NM	-69.102		
	AFM1	-73.792		
	AFM2	-73.673		
	AFM3	-73.608		
	FM	**-74.331**		
H	NM	-83.558	**-84.725**	-84.306
	AFM1	-87.906	**-90.232**	-89.028
	AFM2	-87.367	**-89.677**	-88.631
	AFM3	-87.431	**-89.853**	-88.731
	FM	-88.339	**-90.531**	-89.515
O	NM	**-101.027**	-100.343	-100.625
	AFM1	-103.047	**-105.743**	-104.501
	AFM2	-103.670	**-105.707**	-104.357
	AFM3	-103.535	**-105.725**	-104.667
	FM	-103.528	**-105.531**	-104.678
F	NM	**-91.106**	-90.421	-90.784
	AFM1	-95.511	**-98.653**	-97.289
	AFM2	-98.000	**-96.834**	-136.355
	AFM3	-95.330	**98.100**	-96.759
	FM	-95.951	**-98.847**	-96.925
OH	NM	**-115.539**	-114.487	-114.849
	AFM1	-118.312	**-121.297**	-119.747
	AFM2	-118.342	**-121.263**	-119.592
	AFM3	-118.194	**-120.890**	-120.158
	FM	-118.816	**-121.634**	-120.594

Table S7. The calculated energies (eV) for the most stable model and preferred magnetic orders of $NbCr_2C_2T_x$ (T = H, O, F, OH, or bare).

$NbCr_2C_2$		Model I	Model II	Model III
Bare	NM	-77.758		
	AFM1	-83.387		
	AFM2	-83.170		
	AFM3	-82.957		
	FM	**-84.113**		
H	NM	-91.662	**-93.138**	-92.348
	AFM1	-96.544	**-99.071**	-97.807
	AFM2	-96.302	**-98.619**	-98.195
	AFM3	-96.331	**-98.674**	-97.959
	FM	-97.143	**-99.589**	-98.411
O	NM	-108.300	**-108.524**	-108.208
	AFM1	-111.025	**-114.850**	-113.521
	AFM2	-111.869	**-114.733**	-113.440
	AFM3	-112.277	**-114.731**	-113.512
	FM	-112.085	**-114.910**	-113.592
F	NM	-98.535	**-99.914**	-99.485
	AFM1	-104.984	**-108.016**	-106.395
	AFM2	-104.389	**-107.244**	-106.262
	AFM3	-104.894	**-107.274**	-106.267
	FM	-105.225	**-108.136**	-106.599
OH	NM	**-124.083**	-123.834	-123.816
	AFM1	-128.594	**-131.808**	-130.101
	AFM2	-128.188	**-130.382**	-129.947
	AFM3	-128.145	**-131.220**	-129.662
	FM	-128.875	**-131.595**	-130.615

Table S8. The calculated energies (eV) for the most stable model and preferred magnetic orders of $TaCr_2C_2T_x$ (T = H, O, F, OH, or bare).

$TaCr_2C_2$		Model I	Model II	Model III
Bare	NM	-76.360		
	AFM1	-82.239		
	AFM2	-82.327		
	AFM3	-82.210		
	FM	**-83.011**		
H	NM	-90.543	**-91.938**	-91.173
	AFM1	-95.607	**-98.088**	-96.737
	AFM2	-97.780	**-97.844**	-97.182
	AFM3	-95.661	**-97.930**	-97.056
	FM	-96.381	**-98.755**	-97.575
O	NM	-107.075	**-107.491**	-107.341
	AFM1	-111.462	**-114.559**	-112.907
	AFM2	-111.497	**-114.282**	-112.601
	AFM3	-111.272	**-113.608**	-112.998
	FM	-111.408	**-114.475**	-113.088
F	NM	-98.007	**-98.826**	-98.250
	AFM1	-104.600	**-107.039**	-105.480
	AFM2	-103.871	**-106.521**	-105.801
	AFM3	-106.183	**-106.536**	-105.911
	FM	**-106.982**	-105.031	-105.814
OH	NM	-121.910	**-122.771**	-122.116
	AFM1	-127.418	**-130.536**	-129.418
	AFM2	-127.606	**-130.385**	-129.775
	AFM3	-126.736	**-130.113**	-129.641
	FM	-128.492	**-130.999**	-129.683

Table S9. The calculated energies (eV) for the most stable model and preferred magnetic orders of $MoCr_2C_2T_x$ (T = H, O, F, OH, or bare).

$MoCr_2C_2$		Model I	Model II	Model III
Bare	NM	-77.806		
	AFM1	-82.810		
	AFM2	-82.946		
	AFM3	-82.957		
	FM	**-83.619**		
H	NM	-92.054	**-92.927**	-92.532
	AFM1	-96.432	**-98.905**	-97.659
	AFM2	-96.095	**-98.506**	-97.329
	AFM3	-96.094	**-98.601**	-97.385
	FM	-96.743	**-99.108**	-97.906
O	NM	**-108.850**	-107.795	-108.507
	AFM1	-111.595	**-115.170**	-113.386
	AFM2	-112.144	**-114.015**	-113.539
	AFM3	-112.175	**-114.524**	-113.463
	FM	-112.242	**-115.121**	-113.613
F	NM	**-99.059**	-97.941	-99.604
	AFM1	-107.190	**-107.478**	-105.870
	AFM2	-104.140	**-106.872**	-105.696
	AFM3	-104.010	**-106.833**	-105.264
	FM	-104.685	**-107.350**	-106.158
OH	NM	**-123.550**	-123.380	-123.104
	AFM1	-128.142	**-130.665**	-129.583
	AFM2	-127.365	**-130.030**	-128.049
	AFM3	-127.194	**-130.438**	-127.960
	FM	-126.547	**-130.050**	-129.048

Table S10. The calculated energies (eV) for the most stable model and preferred magnetic orders of $WCr_2C_2T_x$ (T = H, O, F, OH, or bare).

WCr_2C_2		Model I	Model II	Model III
Bare	NM	-81.451		
	AFM1	-86.582		
	AFM2	-86.982		
	AFM3	-87.018		
	FM	**-87.563**		
H	NM	-95.741	**-96.683**	-96.276
	AFM1	-100.307	**-102.845**	-101.562
	AFM2	-100.220	**-102.566**	-101.384
	AFM3	-100.211	**-102.634**	-101.432
	FM	-100.691	**-103.079**	-101.893
O	NM	**-112.487**	-112.418	-111.643
	AFM1	-115.554	**-119.482**	-117.817
	AFM2	-116.367	**-119.193**	-117.973
	AFM3	-116.349	**-119.133**	-117.493
	FM	-116.491	**-119.541**	-117.817
F	NM	-103.079	**-103.445**	-103.249
	AFM1	-110.962	**-111.555**	-109.968
	AFM2	-108.250	**-110.923**	-109.728
	AFM3	-110.003	**-110.861**	-109.528
	FM	-108.696	**-111.474**	-110.034
OH	NM	-127.333	-126.963	**-127.407**
	AFM1	-131.214	**-134.849**	-133.199
	AFM2	-131.289	**-134.180**	-133.375
	AFM3	-131.241	**-133.563**	-132.721
	FM	-132.066	**-134.375**	-133.319