Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information

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Contents

Part 1. The calculated structural and mechanical parameters of Cr_2AlC compared with experimental data.

Table S1. Lattice parameters (in Å), unit cell volumes (in $Å^3$), and the elastic properties (bulk modulus *B*, shear modulus *G*, and Young's modulus *E* (in GPa) of Cr_2AlC studied herein. Also included are previous determinations of the same parameters measured by experiments (Exp.). The NM, NM+U, AFM, and AFM+U represent the results obtained by non-magnetic PBE calculation, PBE+U calculation, calculation with antiferromagnetic spin-ordering, and PBE+U calculation with antiferromagnetic spin-ordering (AFM+U), respectively. The magnetic ordering is in-AFM1.

	NM	NM+U	AFM	AFM+U	Exp.
a	2.841	2.841	2.849	2.864	2.858 [a], 2.86 [b],2.863 [c], 2.854 [d],2.848 [e]
c	12.683	12.683	12.697	12.792	12.808 [a],12.82 [b],12.814 [c], 12.82 [d],12.72 [e]
V	88.653	88.653	89.252	90.869	90.602 [a], 90.814 [b]
B	196.3406	196.3406	179.132	155.008	139 [f],165 [g], 138 [h]
G	117.306	117.306	117.405	113.688	105 [h]
E	293.473	293.473	289.064	274.062	245 [h]

[a]See Ref.[1].

[b]See Ref.[2].

[c]See Ref.[3].

[d]See Ref.[4].

[e]See Ref.[5].

[f]See Ref.[6].

[g]See Ref.[7].

[h]See Ref.[8].

Part 2. The defect formation energies of interstitials with various positions.

Table S2. Defect formation energies (E_{def}, in eV) of Cr_i, Al_i, and C_i in Cr_2AlC. The interstitials are much more stable between Cr-Al layers than between Cr-C layers. The Al_i is unstable between Cr-C layers.

Positions	Cr_i	Al_i	C_i
Cr-Al	4.34	4.49	2.18
Cr-C	6.22	-	4.69

Part 3. The detailed data for defect formation energies in Cr_2AlC.

Table S3. Defect formation energies (E_{def}, in eV) of on-lattice defects in Cr_2AlC. The results of E_{def} for the most studied Ti_3SiC_2 calculated by previous work [9] are also listed for comparison. All the Defect formation energies were calculated with the chemical potentials of atoms assumed to be the bulk crystals of elementary substance.

Defect	NM	AFM+U	Ti_3SiC_2[9]
V_{Cr}	1.95	1.16	5.5
V_{Al}	2.03	2.47	2.1
V_C	0.97	0.81	2.1
Cr_i	4.34	5.42	3.6
Al_i	4.49	4.16	2.1
C_i	2.18	2.24	0.9
Cr_{Al}	1.34	2.13	1.8
Al_{Cr}	0.93	-0.23	3.2
Cr_C	5.32	6.47	8.0
C_{Cr}	4.16	2.92	5.9
Al_C	4.83	4.66	3.4
C_{Al}	15.83	15.65	3.2

Part 4. The projected density of states of Cr_2AlC for Cr-3*d* and C-2*p* orbitals.

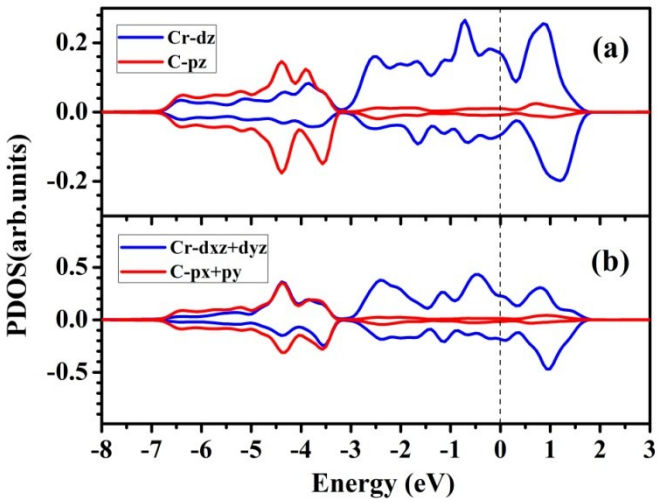

Figure S1. The projected density of states (PDOS) of Cr_2AlC for Cr-d and C-p orbitals. The projection method is the same to Fig. 6, but without the rotation. In other words, the dz^2-orbital is now aligned with the cell axis. It can be seen the dz^2-orbital spin splitting can not be obviously observed without rotation.

Part 5. The detailed data for defect formation energies of Cr_2AlC with different magnetic structures.

Table S4. Defect formation energies (E_{def} in eV, U= 0 eV) of on-lattice defects in Cr_2AlC with different magnetic structures illustrated in Fig. S2.

Defect	NM	(a)	(b)	(c)	(d)	(e)	(f)
V_{Cr}	1.95	1.76	2.03	2.01	2.09	2.08	1.95
V_{Al}	2.03	1.73	2.44	2.24	2.52	2.26	1.77
V_C	0.97	0.71	1.27	1.26	1.37	1.14	0.93
Cr_i	4.34	4.21	4.29	4.29	4.36	4.29	3.80
Al_i	4.49	4.28	4.31	4.44	4.85	4.39	4.40
C_i	2.18	1.97	2.19	2.18	2.26	2.21	2.14
Cr_{Al}	1.34	0.94	1.12	1.19	1.22	1.2	0.72
Al_{Cr}	0.93	0.64	0.92	1.01	1.14	0.92	0.85
Cr_C	5.32	5.16	3.71	3.75	3.85	5.35	3.38
C_{Cr}	4.16	3.99	4.24	4.26	4.3	4.24	3.76
Al_C	4.83	4.58	4.77	4.92	4.69	4.87	4.26
C_{Al}	15.83	15.57	15.76	15.91	15.68	15.86	15.25

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