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Supplementary Information

Superior Electronic Structure of Two-Dimensional 3*d* Transition-Metal Dicarbides: Toward Spintronics

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Table S1. The calculated lattice constant (Å), bond length (Å), thickness h (Å), binding energy E_b (eV), charge transfer (e) between TM and C, and magnetic moments (μ_B) of MC_2 monolayer.

		ScC_2	TiC_2	VC_2	CrC_2	MnC_2	FeC_2	CoC_2	NiC_2	CuC_2	ZnC_2
Lattice constant	a	5.46	5.05	4.75	5.03	5.04	5.01	4.96	4.67	4.98	5.18
	b	3.69	3.48	3.22	3.52	3.53	3.45	3.68	3.18	3.42	3.60
bond I	C-C	1.29	1.32	1.34	1.26	1.26	1.27	1.26	1.31	1.28	1.25
bond II	TM-C(x)	2.26	2.10	1.96	2.12	2.13	2.09	2.11	1.82	1.86	1.96
bond III	TM-C(y)	2.41	2.25	2.16	2.47	2.43	2.40	2.38	2.13	2.29	2.39
h		2.29	2.24	2.24	2.27	2.28	2.25	2.15	1.96	1.61	2.06
E_b		6.31	6.83	6.71	6.55	6.39	6.22	5.81	6.12	6.29	6.45
Charge transfer	TM	-1.70	-1.64	-1.42	-1.22	-1.26	-1.18	-1.06	-0.72	-0.64	-0.50
	C	+0.85	+0.82	+0.71	+0.61	+0.63	+0.59	+0.53	+0.36	+0.32	+0.25
Magnetic moments	TM			1.24	3.83	3.25	3.73	2.69	1.16		
	C			0.04	0.15	0.07	0.08	0.07	0.09		

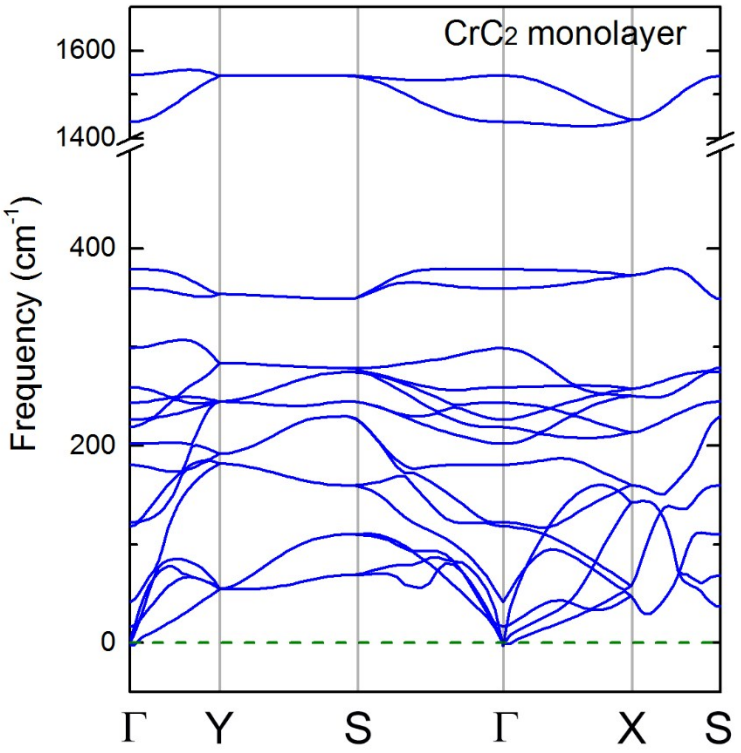

Fig. S1. Phonon bands of monolayer CrC_2 in Brillouin zone.

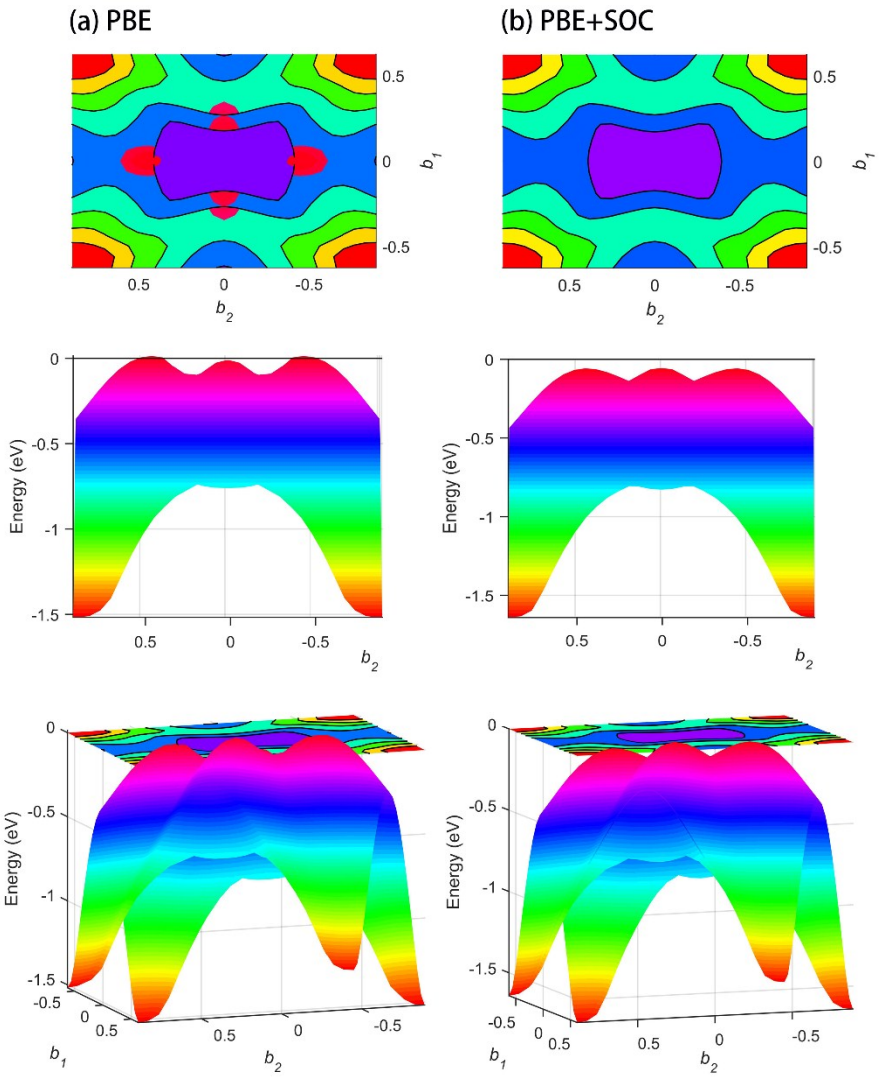

Fig. S2. 3D band structure of monolayer CrC_2 near Fermi level by (a) PBE and (b) PBE+*U*.

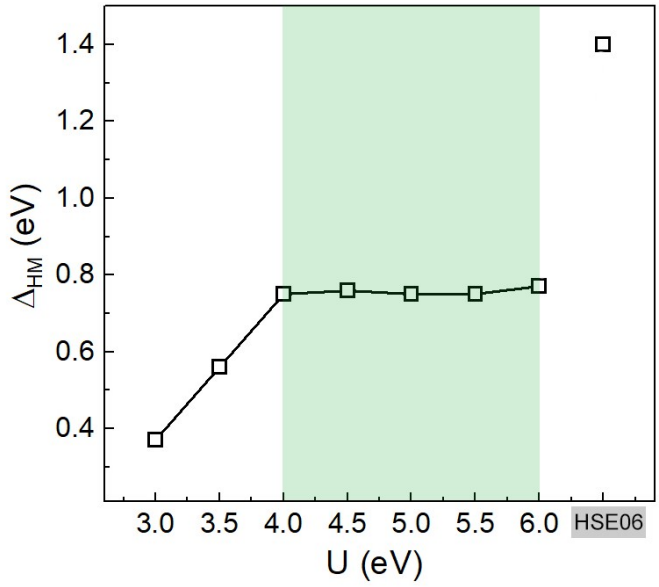

Fig. S3. The energy of CBM in spin-down channel (Δ_{HM}) as a function of U value for half-metallic CrC_2.

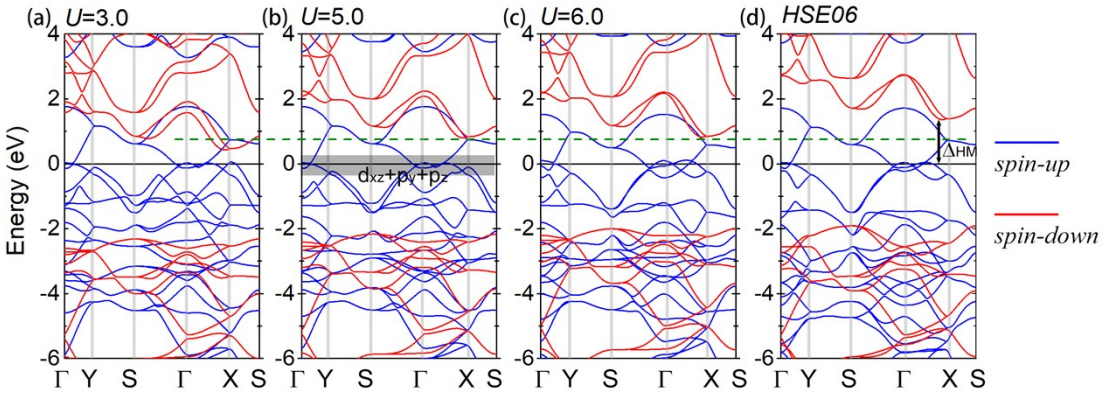

Fig. S4. Band structure of CrC_2 monolayer for different *U* values and HSE06 method. (a) *U* = 3.0 eV, (b) *U* = 5.0 eV, (c) *U* = 6.0 eV, (d) HSE06. The horizontal dashed green line is drawn to guide the eyes. Here, all the structures are relaxed with the different Hubbard *U* values.

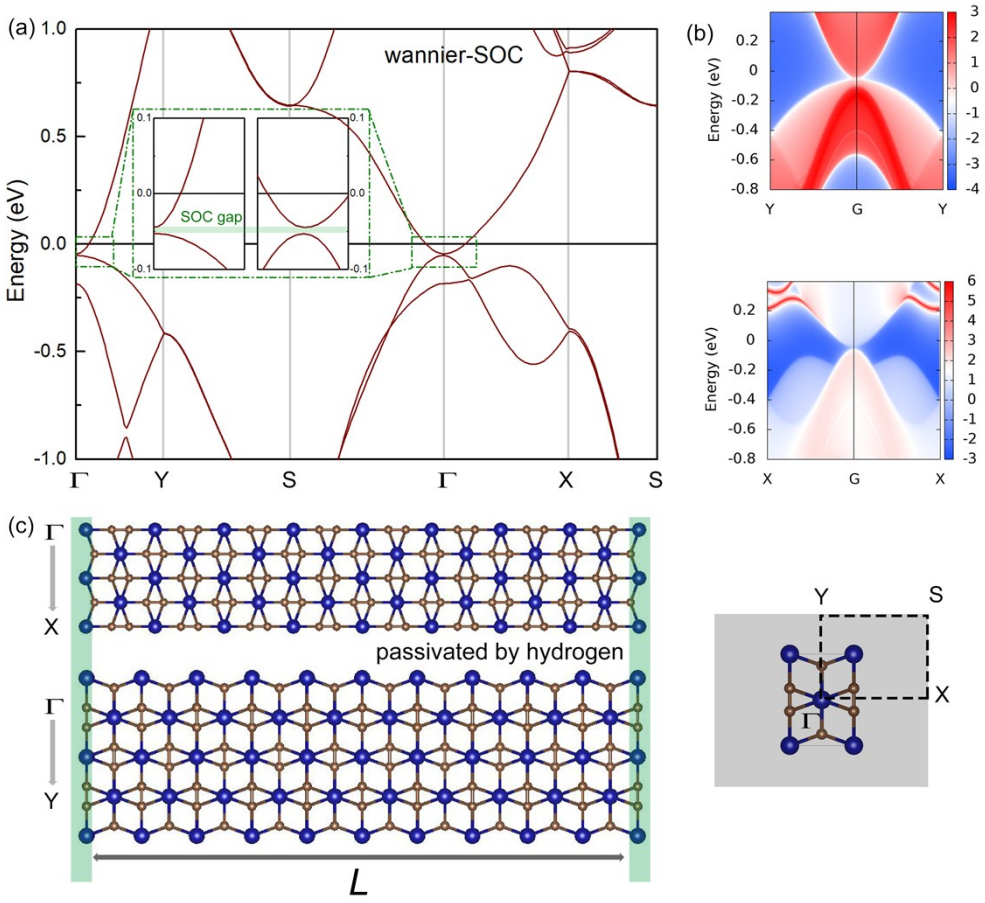

Fig. S5. Band structure of CrC_2 calculated using the MLWFs method with SOC. (b) The chiral edge states of the CrC_2 lattice. (c) Schematic models of the related edge states along Γ-X and Γ-Y directions in Brillouin zone.

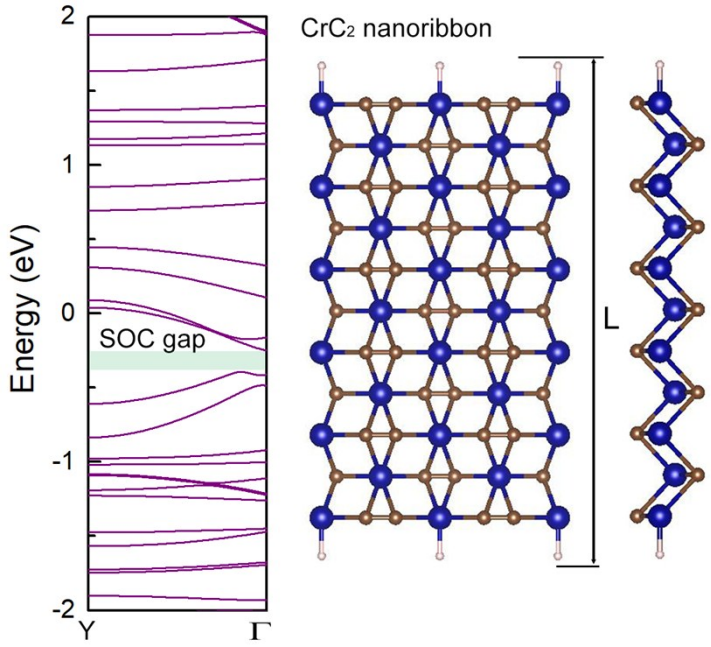

Fig. S6. Calculated electronic band structures of CrC_2 nanoribbon along Γ-Y direction in Brillouin zone with SOC. The Fermi energy is set to 0 eV.

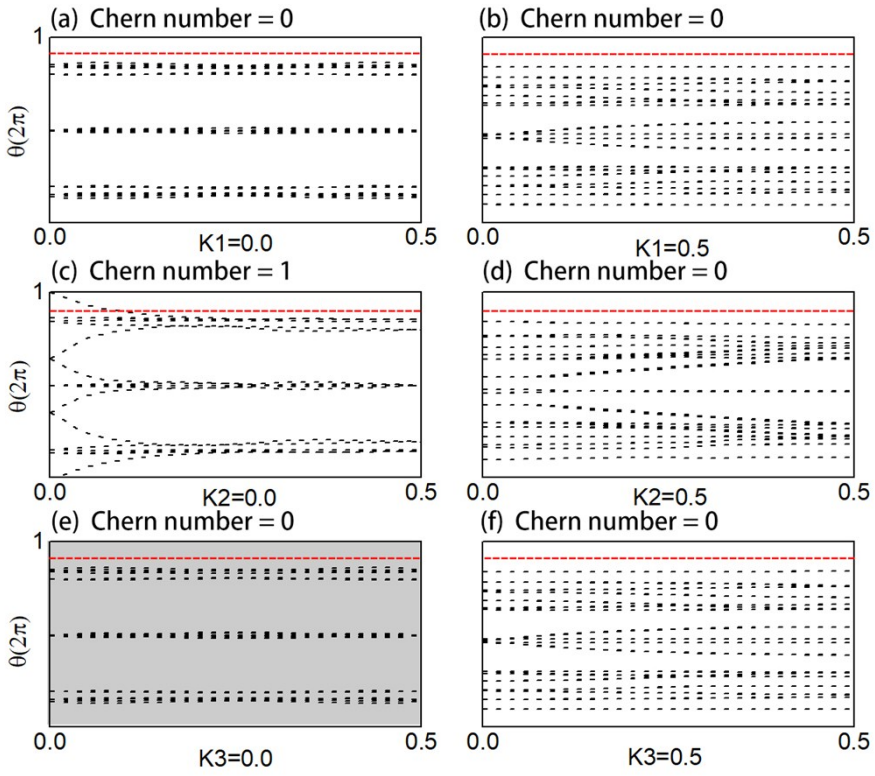

Fig. S7. The evolution lines of Wannier centers of CrC_2 for (a) K1=0.0, (b) K1=0.5, (c) K2=0.0, (d) K2=0.5, (e) K3=0.0, and (f) K3=0.5 planes.