Supplementary Information for

"Rational Design of Two-dimensional Magnetic Chromium Borides from First-principles"

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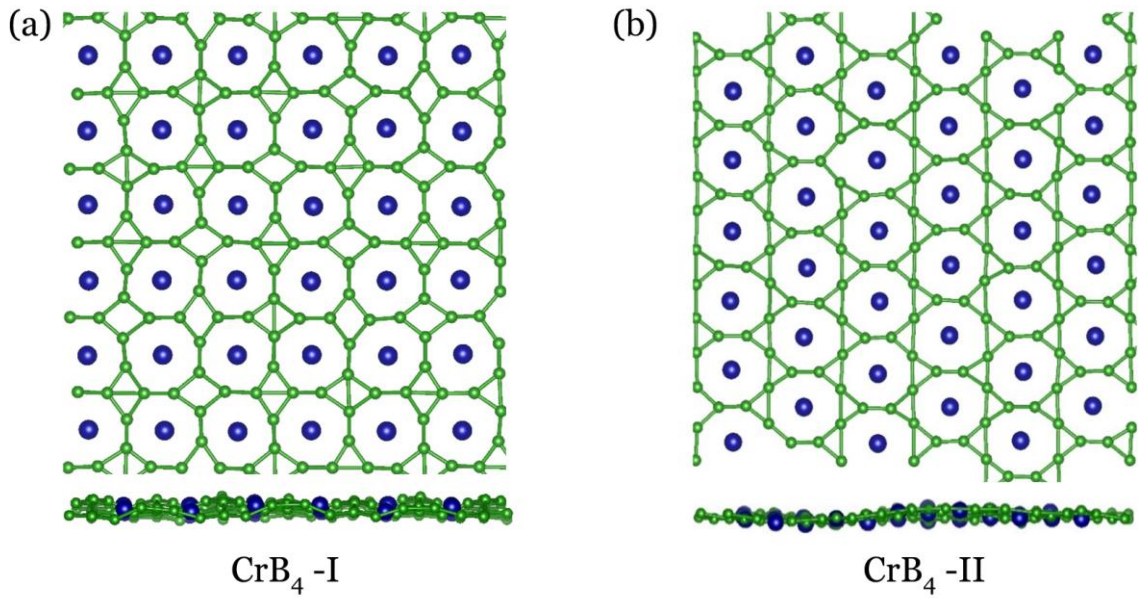

Figure S1. Snapshots of molecular dynamics simulations of (a) CrB_4-I and (b) CrB_4-II, T=300K.

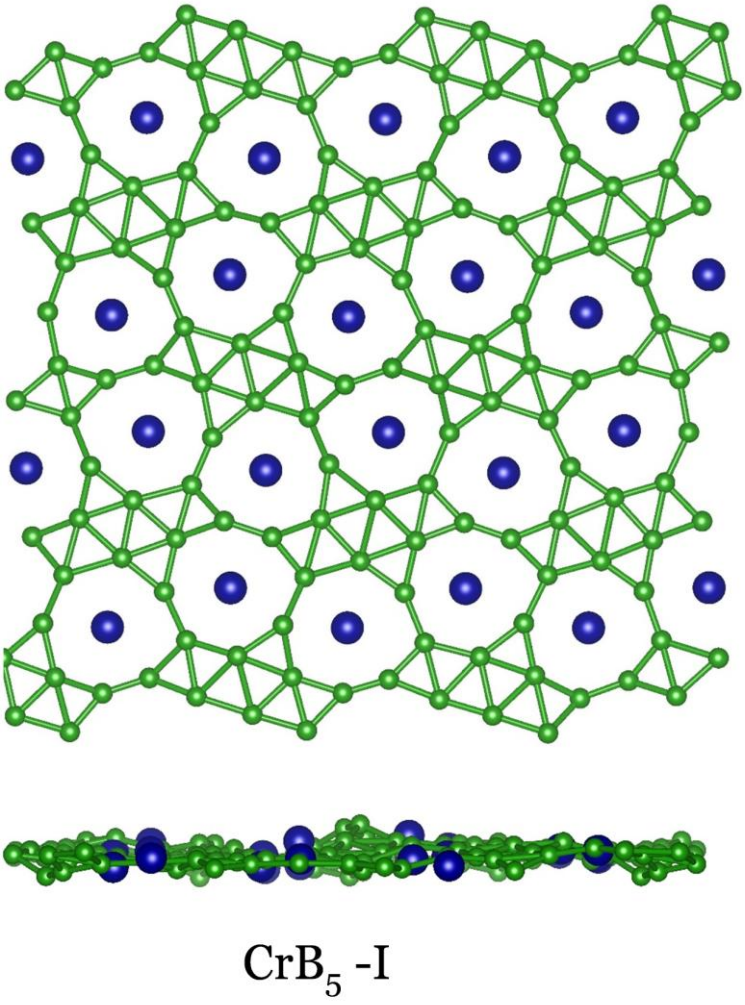

Figure S2. Snapshots of molecular dynamics simulations of CrB_5-I, T=300K.

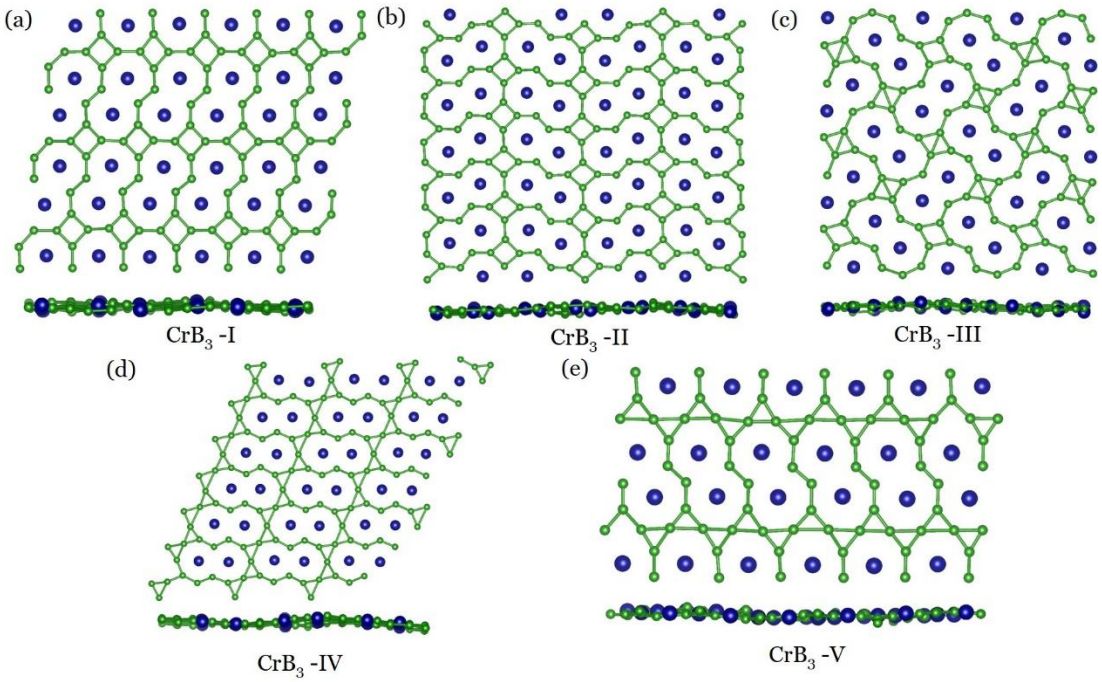

Figure S3. Snapshots of molecular dynamics simulations of CrB_3 series, T=300K. (a)-(e) for CrB_3-I~V, respectively

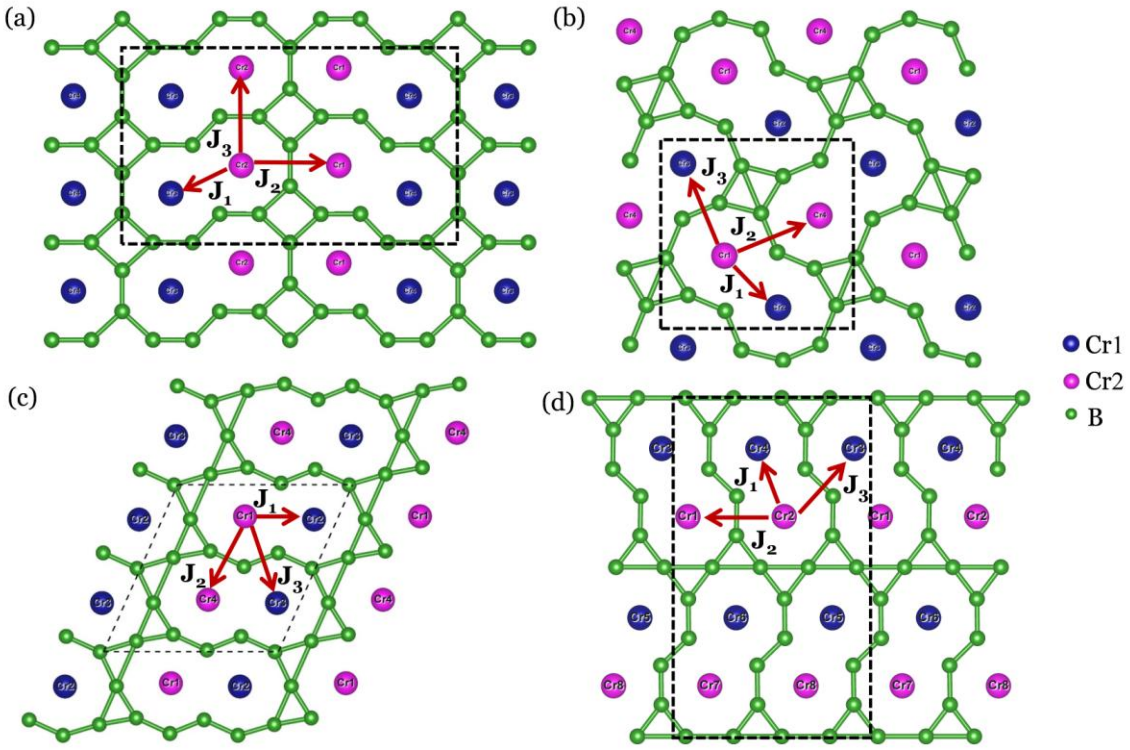

Figure S4. Scheme of antiferromagnetic configurations for (a) CrB_3-II, (b) CrB_3-III, (c) CrB_3-IV and (d) CrB_3-V. Blue and pink balls show the Cr1 and Cr2 atoms with antiparallel spin. The spin of the two Cr atoms in one structural unit is anti-parallel to each other, while for two neighbouring structural units, Cr atoms in corresponding location have same direction of spin. The red vectors mark the directions of calculated magnetic interaction parameters $\boldsymbol{J_1}$, $\boldsymbol{J_2}$ and $\boldsymbol{J_3}$.

		U=3.0eV	U=4.0eV	U=5.0eV
CrB_3-I	J_1 /meV	0.99	1.34	2.34
	J_2 /meV	-6.18	-5.88	-5.35
	J_3 /meV	4.74	4.68	4.64
CrB_3-II	J_1 /meV	8.74	10.61	8.90
	J_2 /meV	-6.75	-7.45	-4.02
	J_3 /meV	-5.51	-6.07	-4.49
CrB_3-III	J_1 /meV	7.06	7.10	7.43
	J_2 /meV	-2.43	-2.07	-1.34
	J_3 /meV	3.53	3.55	3.71
CrB_3-IV	J_1 /meV	13.36	10.77	14.48
	J_2 /meV	-9.37	-6.98	-7.01
	J_3 /meV	4.38	2.84	5.63
CrB_3-V	J_1 /meV	6.44	7.38	9.54
	J_2 /meV	-8.56	-6.89	-4.49
	J_3 /meV	-1.19	-0.78	-0.23
CrB_2-I	J_1 /meV	-2.24	-1.93	-2.22
	J_2 /meV	3.44	3.73	3.94

Table S1. The magnetic exchange parameters J_1, J_2 and J_3 of CrB_3-II/III/IV/V and CrB_2-I under various PBE+U method with Hubbard-U of 3.0, 4.0 and 5.0 eV, derived from energy mapping analysis.

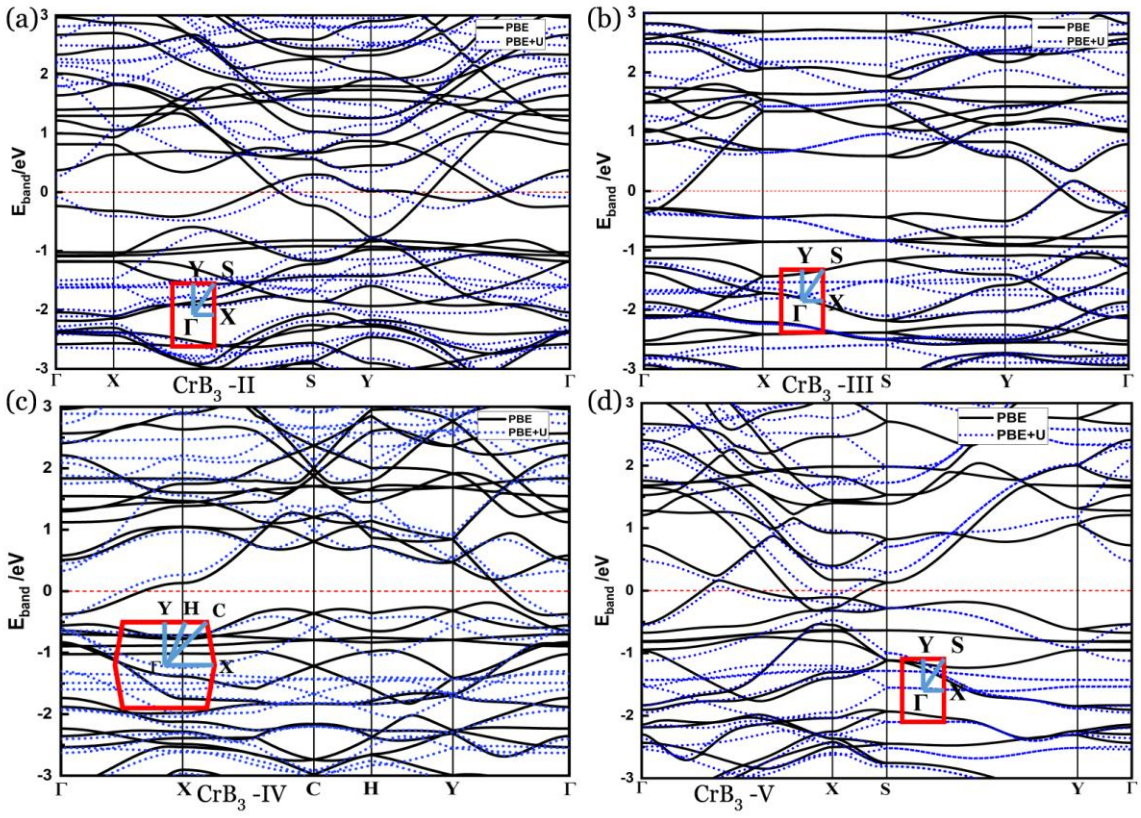

Figure S5. The band structures of AFM CrB_3-II/III/IV/V. The black solid lines show the PBE band, the blue dotted lines show the PBE+U band. The red dotted line shows the Fermi energy level, and the high symmetric point path in the Brillouin region is shown internally.

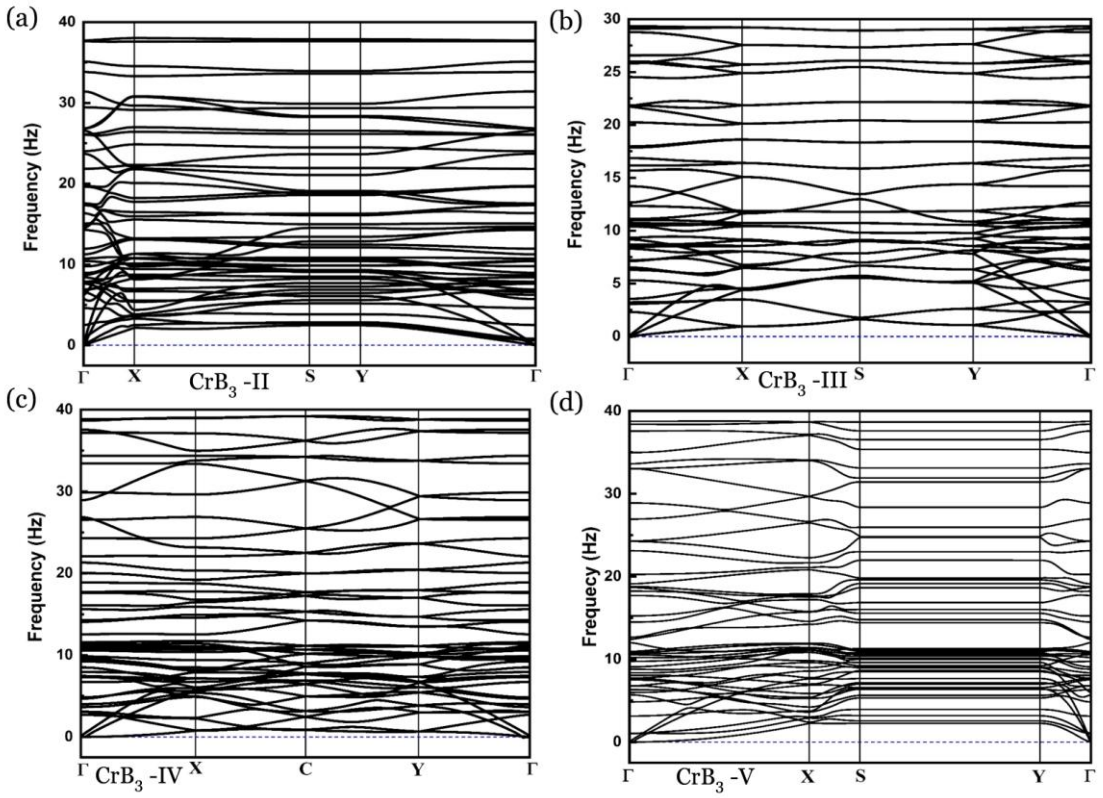

Figure S6. The phonon spectra of AFM CrB_3-II/III/IV/V.