Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Half-metallicity and Enhanced Ferromagnetism in Li-adsorbed Ultrathin Chromium Triiodide

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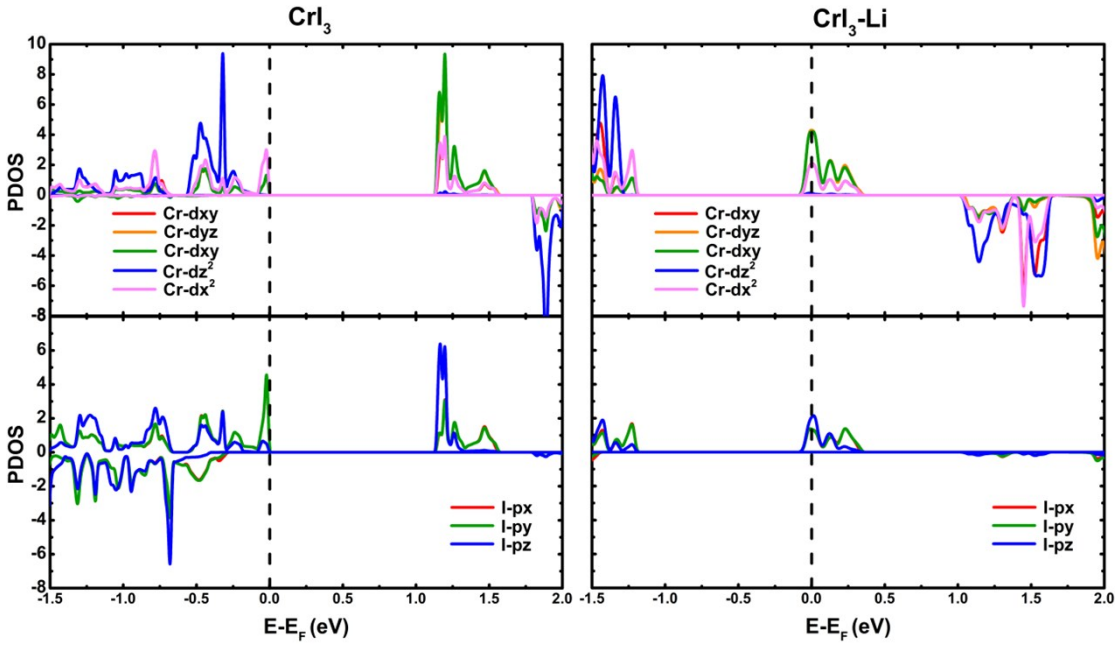

Figure S1: Projected density of states (PDOS) of Cr 3*d* and I 5*p* orbitals, for (a) CrI_3 and (b) CrI_3-Li monolayer, respectively.

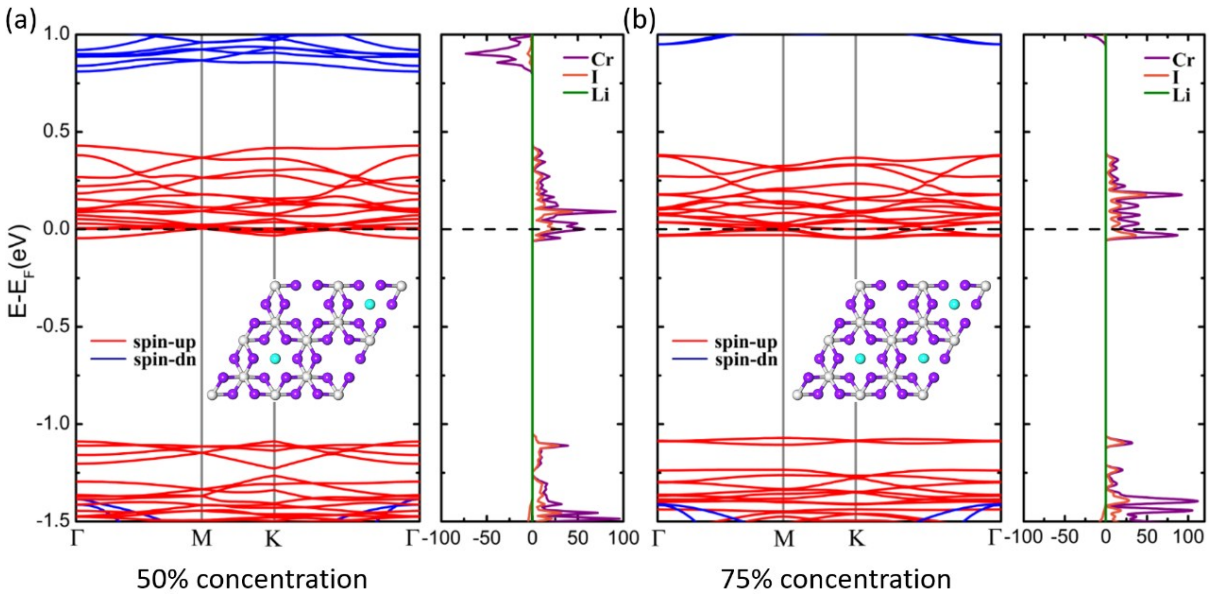

Figure S2: Band structure and PDOS of (a) 1/2 and (b) 3/4 occupied Li-CrI_3. The inset are the structures of 1/2 and 3/4 occupied Li-CrI_3, respectively.

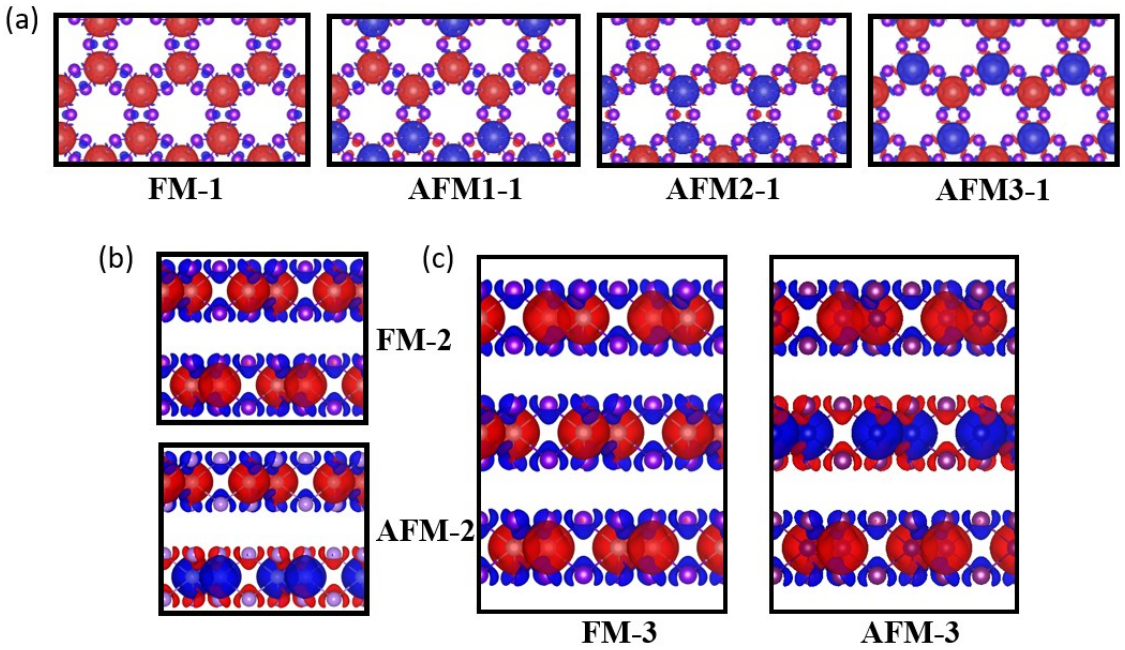

Figure S3: Spin charge density map of different (a) in-plain spin orderings between Cr atoms in monolayer CrI_3 and (b-c) out-of-plain spin orderings in the bi- and tri-layer CrI_3. The red and blue isovalues are the majority and minority spin density of 0.004 e/Å^3 and 0.003 e/Å^3 for monolayer and multi-layer respectively.

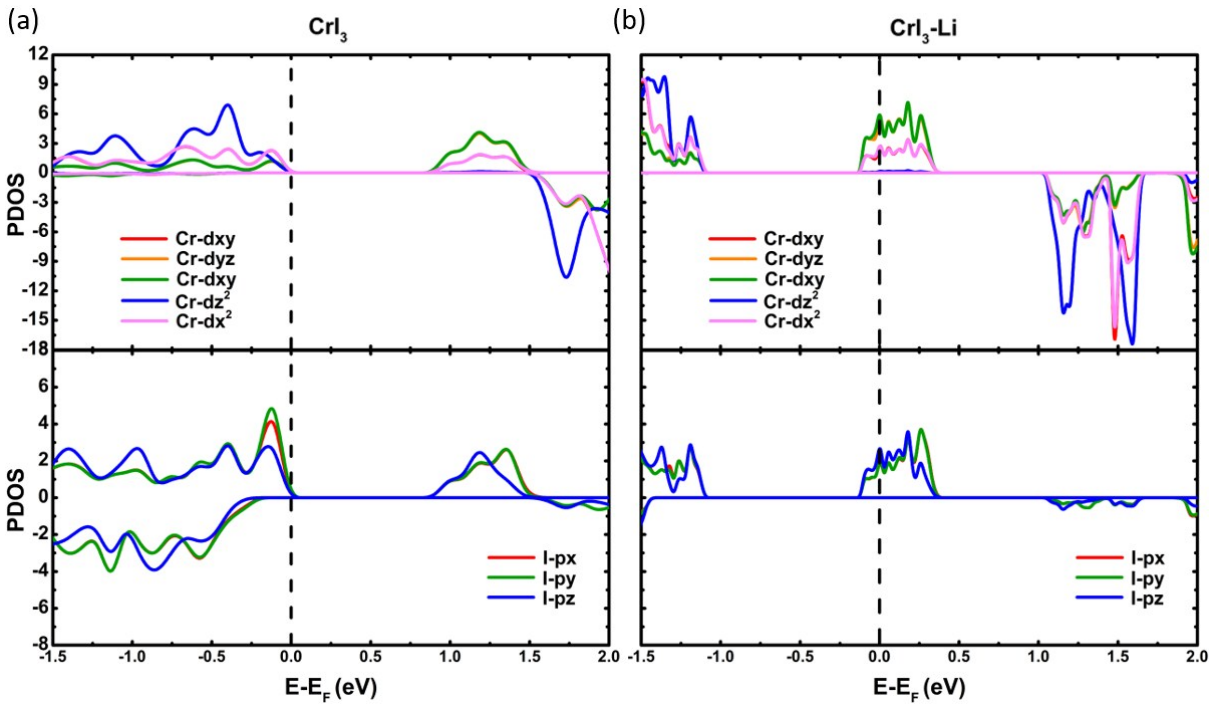

Figure S4: PDOS of Cr 3*d* and I 5*p* orbitals, for (a) CrI_3 and (b) CrI_3-Li trilayer, respectively.

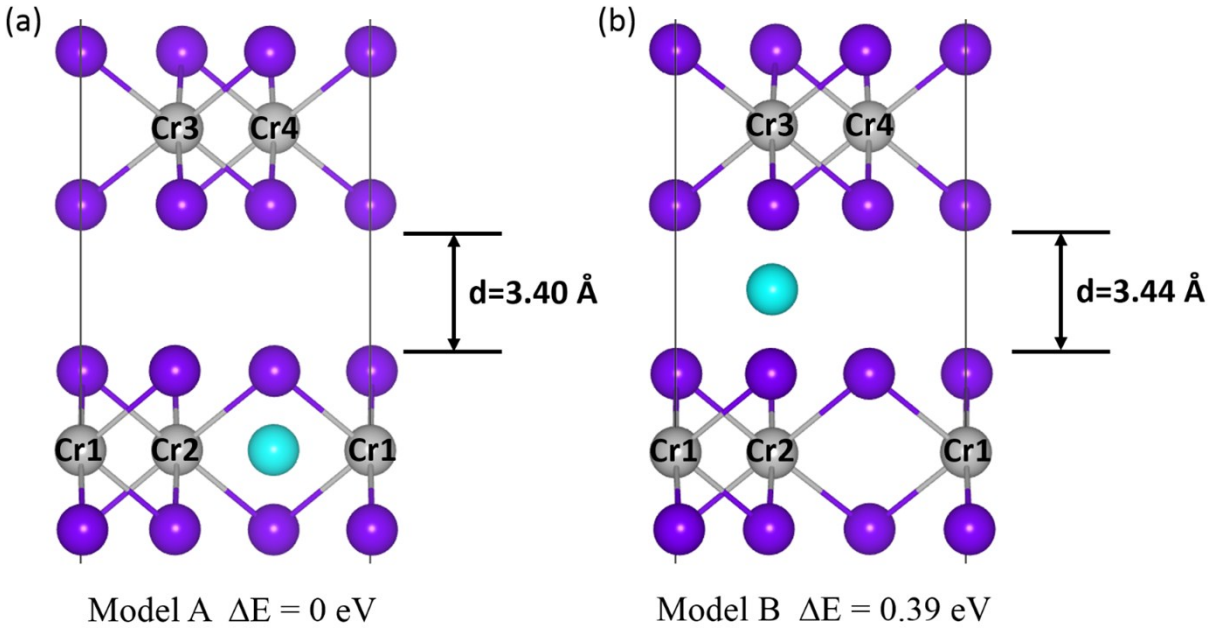

Figure S5: The optimized geometric structures, interlayer distances and the relative total energies of the two different structures of one Li atom in CrI_3 bilayer.

Table S1: Bader charge analysis. Q(Cr) and Q(I) refer to the net charge of Cr and I atoms in CrI_3/CrI_3-Li unit cell, respectively. ΔQ(Cr) and ΔQ(I) refer to the change in charge before and after Li doping.

Layer	CrI_3		CrI_3-Li			ΔQ(Cr)	ΔQ(I)
	Q (Cr)	Q(I)	Q(Cr)	Q(I)	Q(Li)		
1ML	-2.11	2.11	-2.06	2.94	-0.88	0.05	0.83
2ML	-4.21	4.21	-4.12	5.87	-1.75	0.09	1.66
3ML	-6.33	6.33	-6.17	8.80	-2.63	0.16	2.47

Table S2: Total energies of different spin orderings for CrI_3 and CrI_3-Li with 1-3 layers. E_{AFM-FM} is the energy difference between the most stable AFM and FM spin orderings. The most stable AFM states are highlighted in bold-typeface.

layers	Unit cell	E_{FM} (eV)	E_{AFM1} (eV)	E_{AFM2} (eV)	E_{AFM3} (eV)	E_{AFM-FM} (meV)
1L	CrI_3	-31.566	**-31.545**	-31.529	-31.530	21
	CrI_3-Li	-35.069	**-35.038**	-35.036	-35.002	31
2L	CrI_3	-67.146	-67.134	-	-	12
	CrI_3-Li	-75.310	-75.228	-	-	82
3L	CrI_3	-101.058	-101.034	-	-	24
	CrI_3-Li	-113.310	-113.159	-	-	151

Table S3: Magnetic moments of Cr atoms of the Model A and Model B structures shown in Figure S5.

Atom number	M_A (μ_B)	M_B (μ_B)
1	3.34	3.21
2	3.39	3.21
3	3.07	3.21
4	3.04	3.21
Average	3.21	3.21