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Supplementary Information for

High Curie Temperature and Intrinsic Ferromagnetic Half-Metallicity in Two-Dimensional Cr_3X_4 (X= S, Se, Te) Nanosheets

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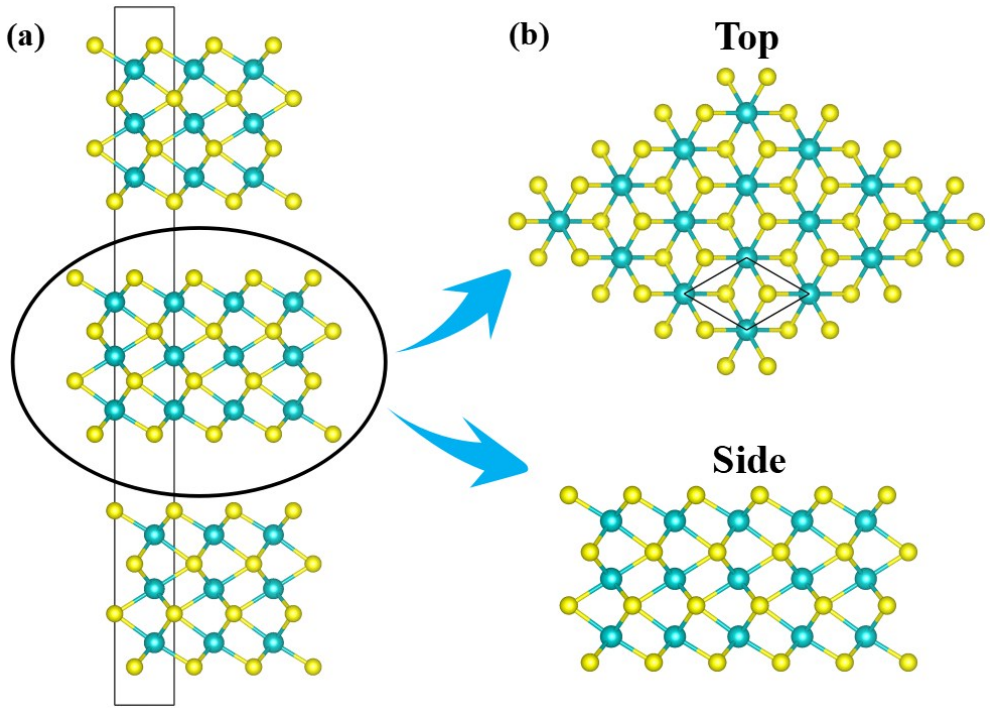


Fig. S1 (a) Crystal structure of layered Fe_3S_4 bulk. (b) Top and side views of one Fe_3S_4 sheet. The green and yellow balls represent Fe and S atoms.

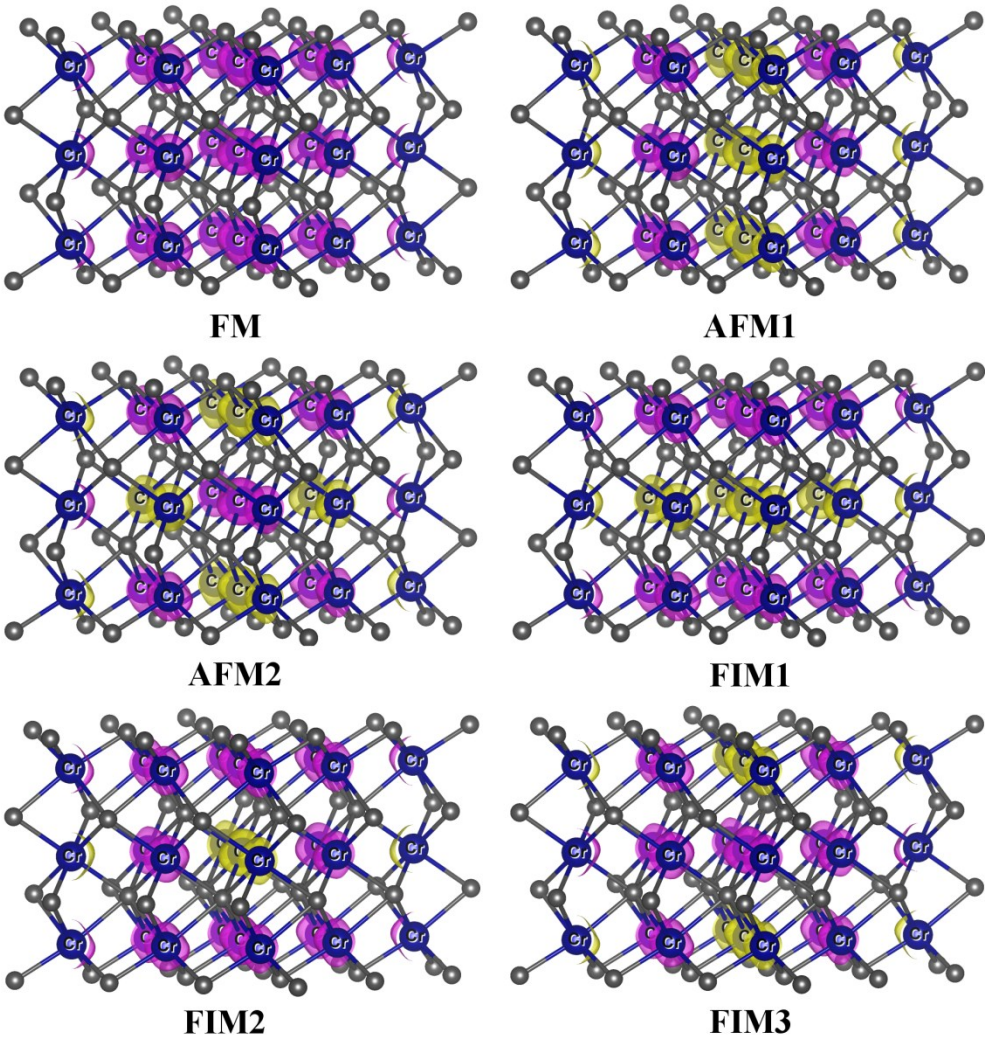


Fig. S2 Possible magnetic configurations and the corresponding spin charge densities of Cr_3X_4 (X= S, Se, Te) monolayers.

Table S1 Relative energies (meV/f.u.) between ferromagnetic (FM) and antiferromagnetic/ferrimagnetic (AFM1, AFM2, FIM1, FIM2, and FIM3) states for Cr_3X_4 (X= S, Se, Te) monolayers. Ground states are highlighted with green backgrounds.

Material Name	E_{FM-FM}	$E_{FM-AFM1}$	$E_{FM-AFM2}$	$E_{FM-FIM1}$	$E_{FM-FIM2}$	$E_{FM-FIM3}$
Cr_3S_4	0	-78.5	45.7	146.6	70.4	-27.8
Cr_3Se_4	0	-292.9	-225.2	-145.5	-58.6	-268.7
Cr_3Te_4	0	-487.6	-456.9	-367.3	-218.6	-350.2

Table S2 Summary of magnetocrystalline anisotropy energy in μeV/Cr for Cr_3Se_4 and Cr_3Te_4 monolayers.

Material Name	E(100) – E(001)	E(010) – E(001)	E(001) – E(001)
Cr_3Se_4	145.9	146.0	0
Cr_3Te_4	101.0	127.3	0

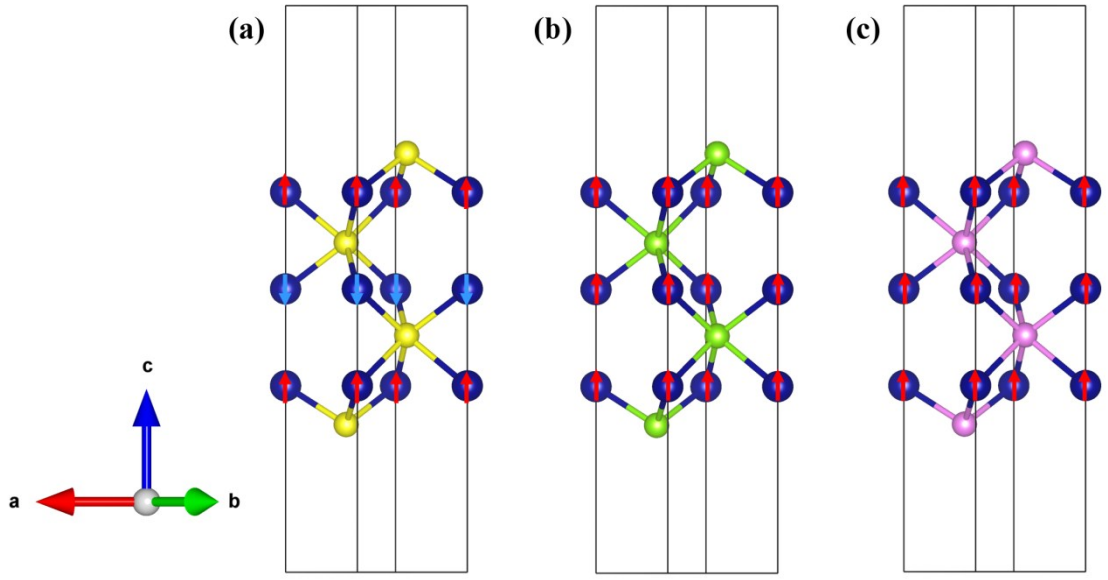


Fig. S3 Ground-state spin configurations in the unit-cell of Cr_3X_4 (X= S, Se, Te) monolayers.

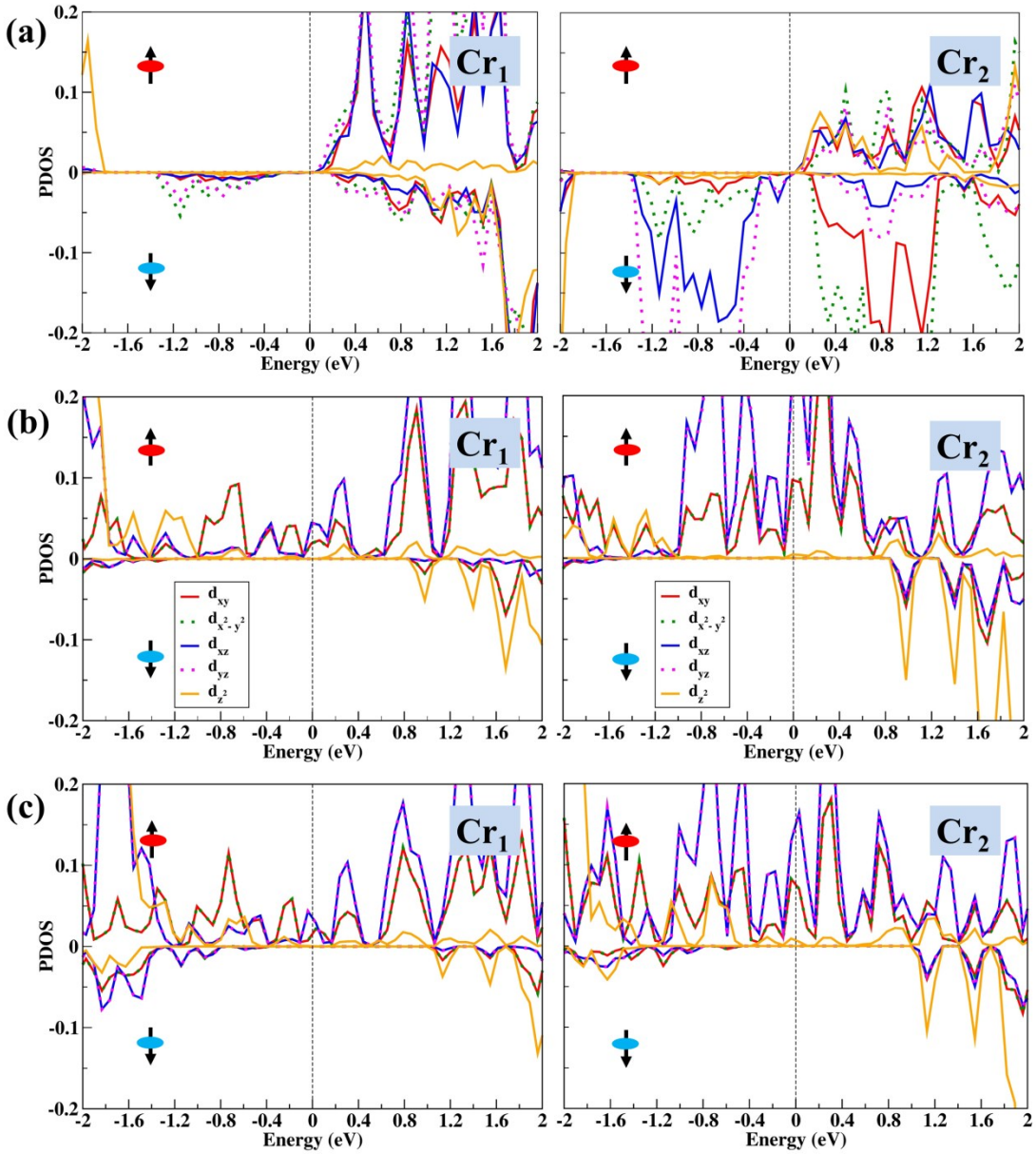
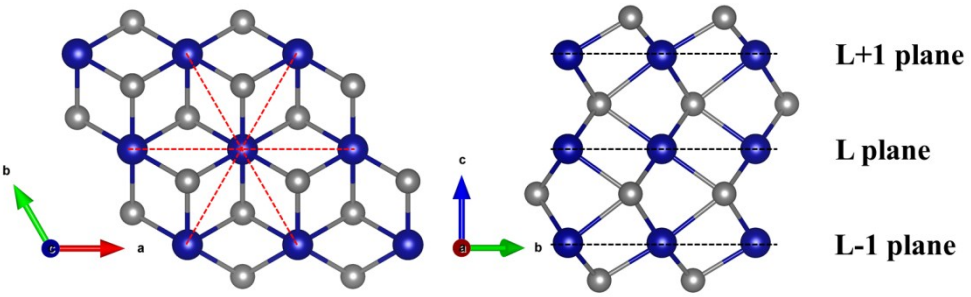


Fig. S4 Calculated partial density of states (PDOS) of (a) Cr_3S_4, (b) Cr_3Se_4, and (c) Cr_3Te_4 monolayers.

Table S3 Lattice constant (a_0), Cr_1-X-Cr_1 (θ) and Cr-Cr/X bond length of single-layer Cr_3X_4 (X= S, Se and Te).

Materials	a_0 (Å)	Θ (deg)	Cr_1-Cr_1 (Å)	Cr_1-X_1 (Å)	Cr_2-X_2 (Å)	Cr_1-Cr_2 (Å)
Cr_3S_4	3.44	95.90	3.44	2.32	2.43	3.02
Cr_3Se_4	3.68	95.72	3.68	2.48	2.62	3.19
Cr_3Te_4	4.01	95.63	4.01	2.71	2.85	3.26



$$H = -J_1 \sum_{ill'} \delta_{|l-l'|,1} S_{l,i} S_{l',i} - J_2 \sum_{il\sigma} \delta_{|l-L|,1} S_{l,i} S_{l,i+\sigma} - J_3 \sum_{il\sigma} \delta_{|l-L|,0} S_{l,i} S_{l,i+\sigma}$$

$$l, l' = L-1, L, L+1$$

Fig. S5 Top and side views of magnetic structure for the spin Hamiltonian of Cr_3Se_4 and Cr_3Te_4 monolayers. J_1 and J_2/J_3 are defined as interlayer and intralayer exchange coupling parameters. A is anisotropy energy parameter. i and $i+\sigma$ respect the positions of the two interacting Cr atoms. $S^z_{l,i}$ is the spin vector of each Cr atom. Dark blue and gray balls respect Cr and X atoms, respectively.

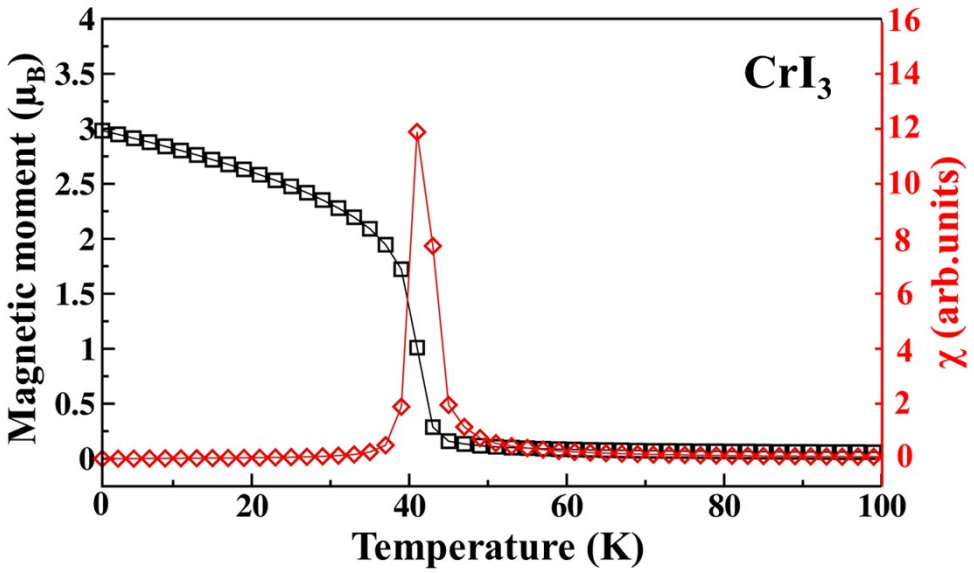


Fig. S6 Magnetic moment (black data) and magnetic susceptibility χ (red data) as functions of temperature for CrI_3 monolayers by Monte Carlo simulations on the basis of 2D Heisenberg Hamiltonian model.

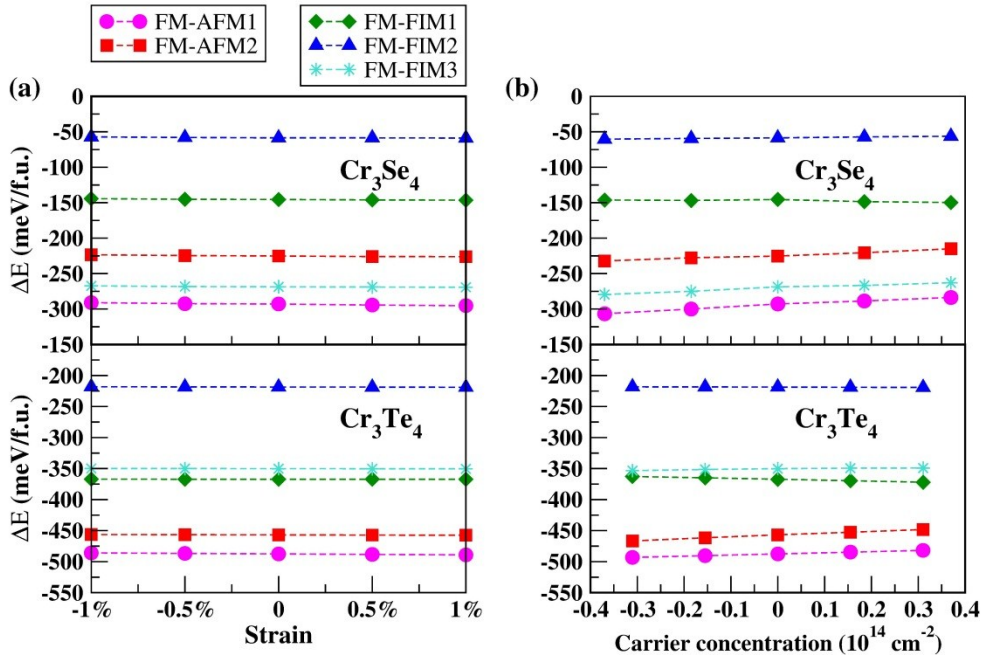


Fig. S7 Relative energies of FM/AFM/FIM states as a function of strain (a) and carrier concentration (b) for 2D Cr_3Se_4 and Cr_3Te_4. The positive and negative values refer to electron and hole doping, respectively.