Supporting Information for

Chromium Sulfide Halide Monolayers: Intrinsic Ferromagnetic Semiconductors with Large Spin Polarization and High Mobility

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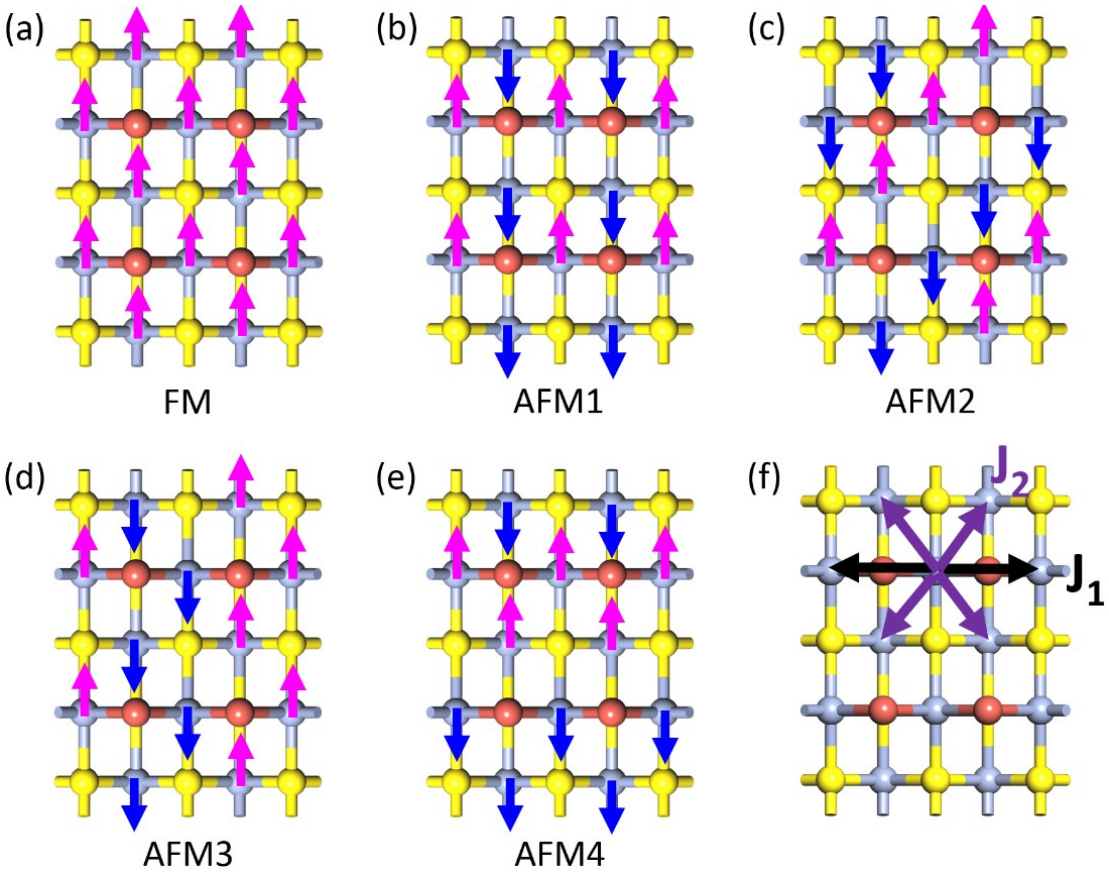

Figure S1. (a-e) Top views of geometric structures for ferromagnetic and four different antiferromagnetic states for 2D CrSX monolayers. Pink and blue arrows denote spin-up and spin-down orientations on Cr atoms, respectively. (f) The nearest- and next-nearest-neighbor exchange interactions. J_1 and J_2 represent the first and second nearest-neighboring exchange parameters,

respectively.

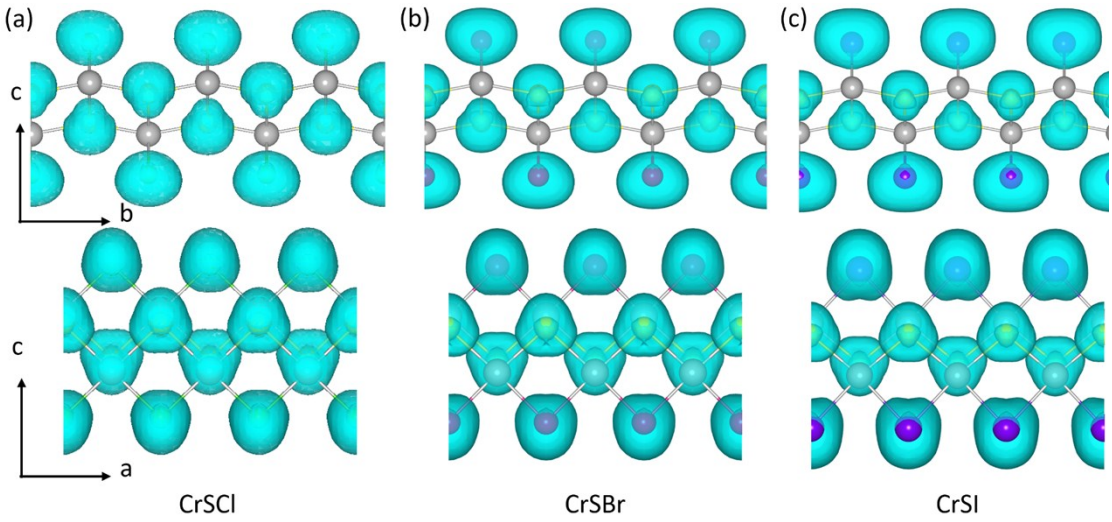

Figure S2. Side view of electron localization functions of (a) CrSCl, (b) CrSBr and (c) CrSI monolayers with an isosurface of 0.5 along a and b directions.

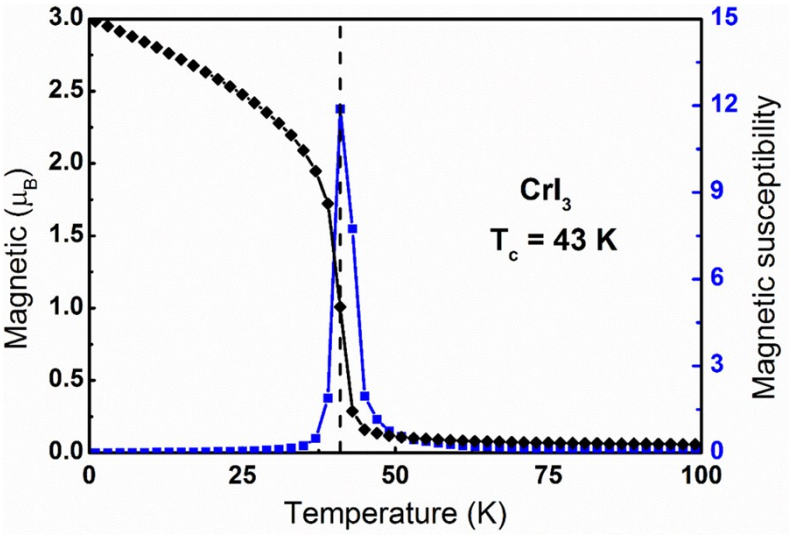

Figure S3. The simulated magnetic moment (black data) and magnetic susceptibility (blue data) as functions of temperature for CrI_3 monolayer.

To confirm the reliability of the method for the prediction of Curie temperature, we calculated the Tc for the single-layer CrI_3. The value of the nearest exchange interaction constant and the single-ion anisotropy energy parameter are 2.67 meV and

825 μeV/Cr, respectively, which is consistent with the values (2.71 meV, 686μeV/Cr) in Ref. 1. The calculated Tc for CrI_3 monolayer is about 43 K, which is greatly close to the experimental measurement of 45 K from Ref. 2. This indicates the reliability of our method.

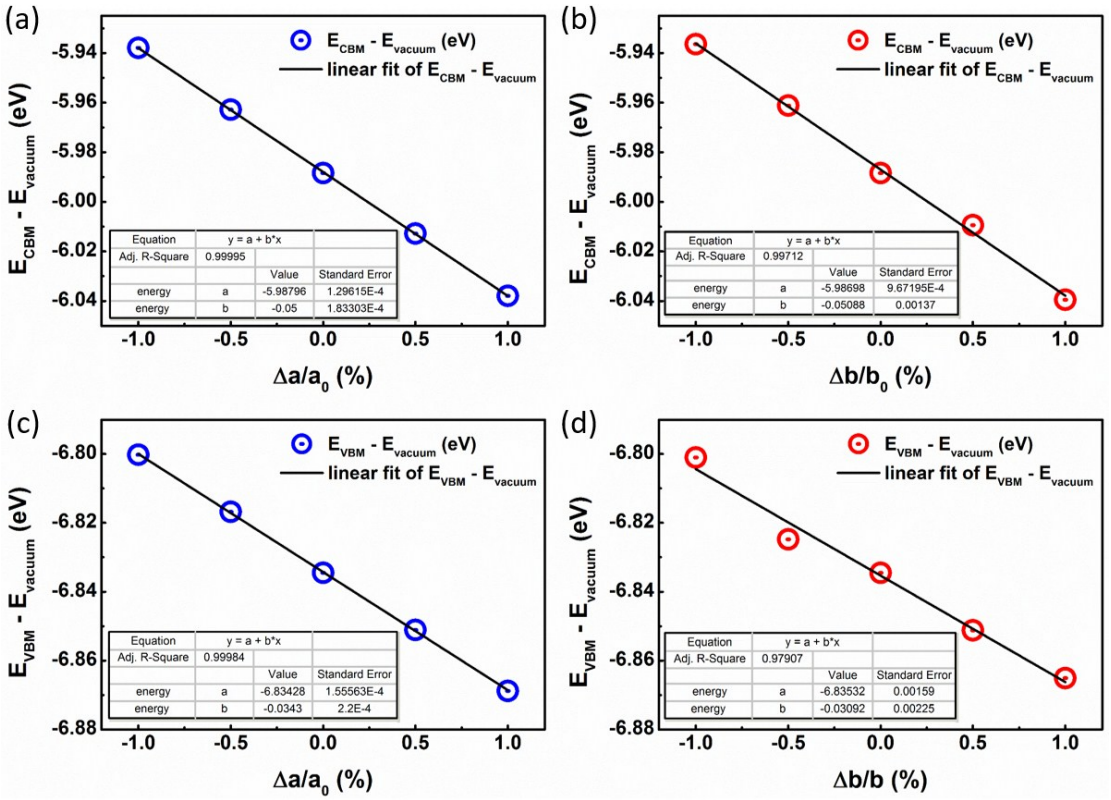

Figure S4. Band energy with respect to the vacuum energy as a function of lattice dilation for the (a, b) CBM and (c, d) VBM of CrSCl monolayer. Black solid lines are the fitting curves. The standard errors are shown Inset for the fitted slope, corresponding to the deformation potential.

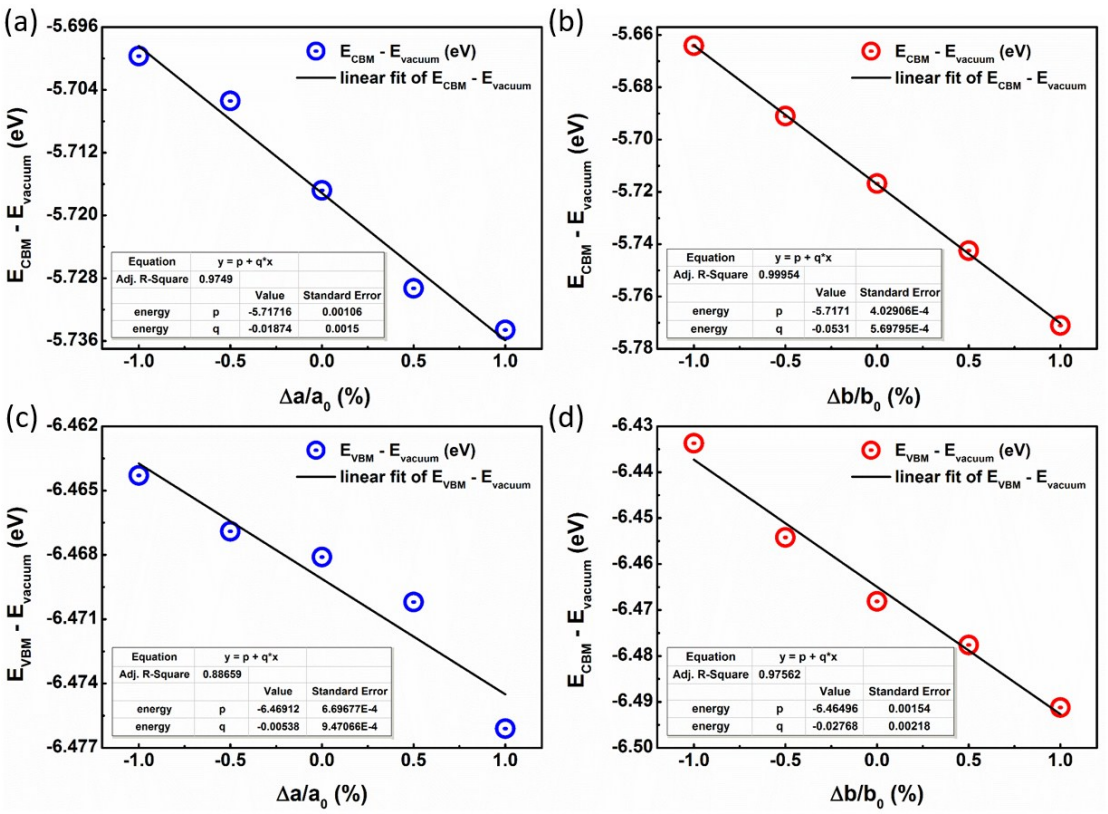

Figure S5. Band energy with respect to the vacuum energy as a function of lattice dilation for the (a, b) CBM and (c, d) VBM of CrSBr monolayer. Black solid lines are the fitting curves. The standard errors are shown Inset for the fitted slope, corresponding to the deformation potential.

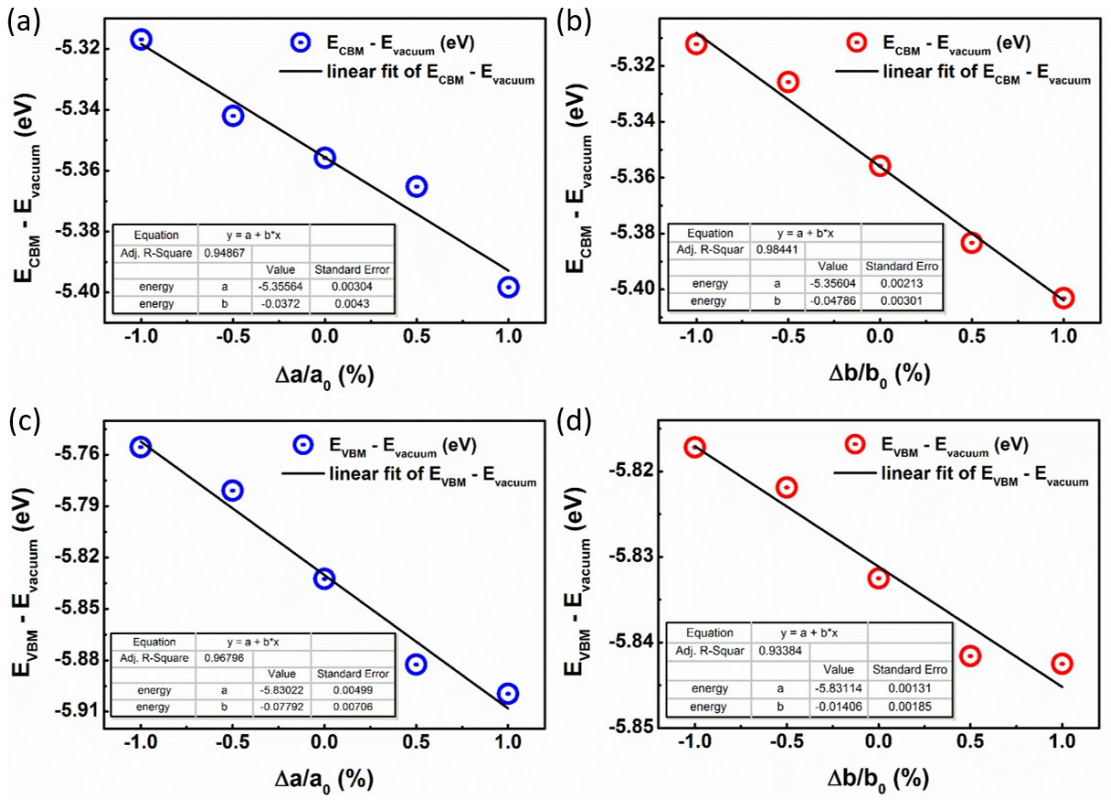

Figure S6. Band energy with respect to the vacuum energy as a function of lattice dilation for the (a, b) CBM and (c, d) VBM of CrSI monolayer. Black solid lines are the fitting curves. The standard errors are shown Inset for the fitted slope, corresponding to the deformation potential.

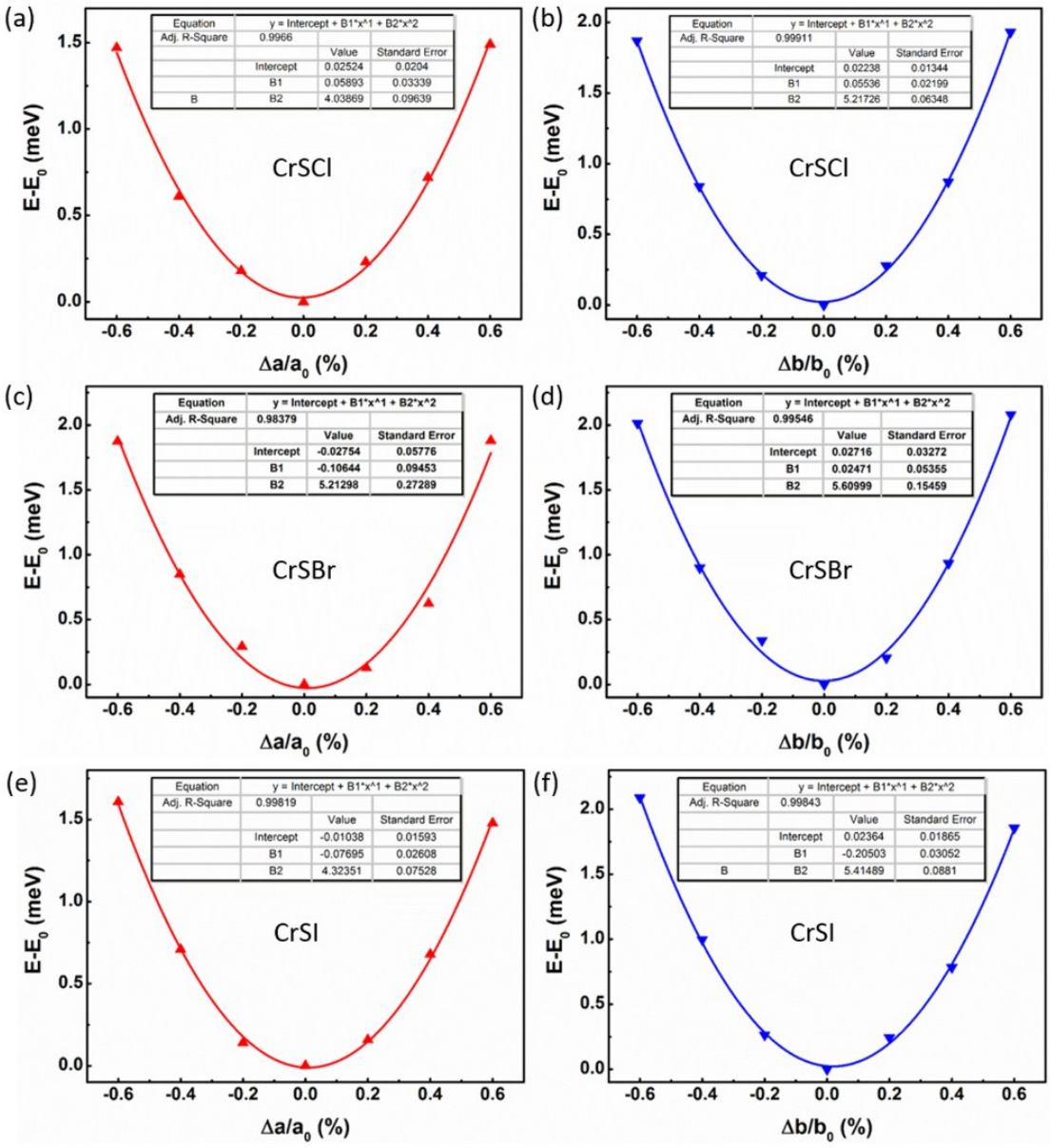

Figure S7. Total energy-strain curve of CrSX monolayers along the (a, c and e) a and (b, d and f) b directions, respectively. Insets show the standard errors for the fitted hyperbolic curves.

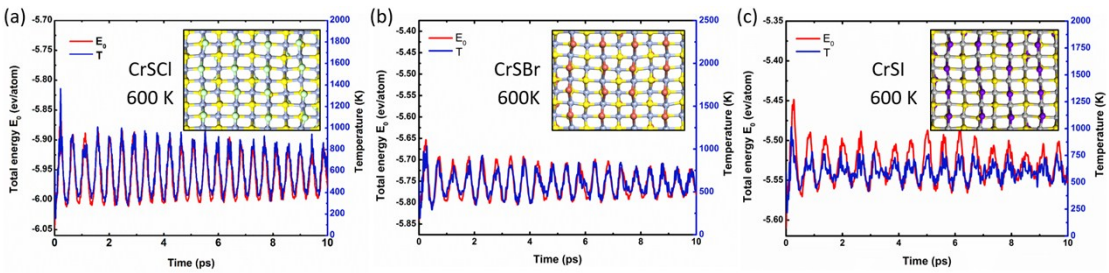

Figure S8. AIMD evolutions of total energy and temperature for CrSCl, CrSBr and CrSI monolayers at 600 K, respectively. The insets show the snpshots of the associated monolayers at 10 ps.

Table S1. Optimized lattice constant a and b (Å), selected bond lengths (Å) and angles (˚), electronic band gap Eg (eV) of CrSX monolayer. Experimental values for bulk CrSBr are given for comparison.

Name		**a**	**b**	**c**	***l*1**	***l*2**	***l*3**	**θ1**	**θ2**	**θ3**	**Eg**
CrSCl	1 ML	3.49	4.84	-	2.40	2.41	2.46	92.52	93.28	97.05	0.856
CrSBr	1 ML	3.59	4.83	-	2.54	2.43	2.45	95.35	89.63	96.99	0.757
	bulk	3.55	4.80	7.98	-	-	-	-	-	-	-
	exp	3.51	4.77	7.97	-	-	-	-	-	-	-
CrSI	1ML	3.76	4.81	-	2.75	2.45	2.45	100.05	86.28	96.95	0.473

Reference

1. W.-B. Zhang, Q. Qu, P. Zhu and C.-H. Lam, *J. Mater. Chem. C,* 2015, **3**, 12457.

2. B. Huang, G. Clark, E. Navarro-Moratalla, D. R. Klein, R. Cheng, K. L. Seyler, D. Zhong, E. Schmidgall, M. A. McGuire, D. H. Cobden, W. Yao, D. Xiao, P. Jarillo-Herrero and X. Xu, *Nature,* 2017, **546**, 270.