

# Supplemental Material Information

## “Hole-doping-induced Perpendicular Magnetic Anisotropy and Room-Temperature Ferromagnetism in Monolayer CrSX (X=Cl, Br, I)”

Ruilin Han,<sup>\*a</sup> Xiaomin Xue,<sup>b</sup> and Yu Yan<sup>c</sup>

<sup>a</sup> *School of Physics and Electronic Engineering, Shanxi University, Taiyuan 030006, China*

<sup>b</sup> *Institute of Theoretical Physics, Shanxi University, Taiyuan, 030006, China*

<sup>c</sup> *Key Laboratory of Physics and Technology for Advanced Batteries (Ministry of Education),  
Department of Physics, Jilin University, Changchun 130012, China*

<sup>\*</sup>E-mail: hanruilin0116@sxu.edu.cn

**Table S1.** Calculated in-plane lattice constants, bond lengths, bond angles and band gaps of the Janus CrSX (X=Cl, Br, I) monolayers.

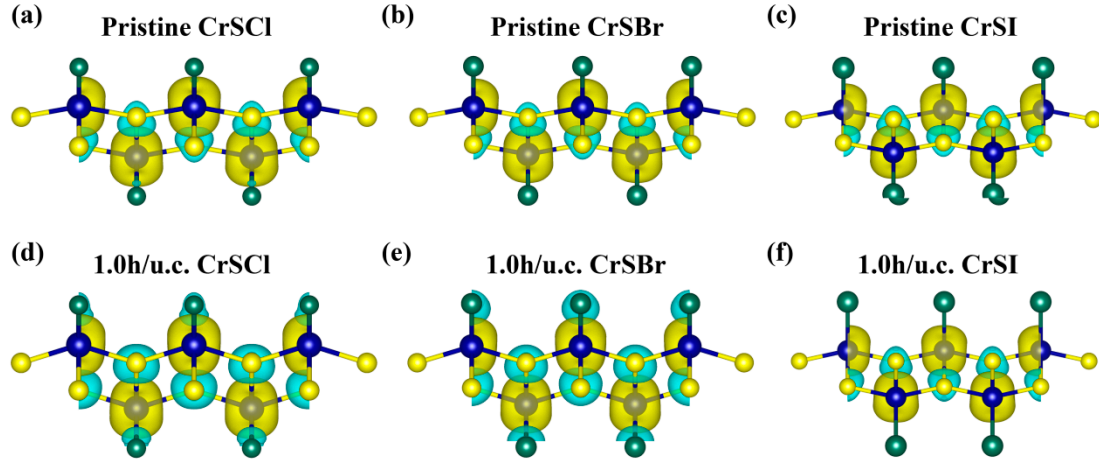
| Pristine | a(Å) | b(Å) | $\theta_1(^{\circ})$ | $\theta_2(^{\circ})$ | $\theta_3(^{\circ})$ | $l_1(\text{Å})$ | $l_2(\text{Å})$ | $l_3(\text{Å})$ | band-gap(eV) |
|----------|------|------|----------------------|----------------------|----------------------|-----------------|-----------------|-----------------|--------------|
| CrSCl    | 3.48 | 4.83 | 93.21                | 92.44                | 97.03                | 2.39            | 2.41            | 3.48            | 1.78         |
| CrSBr    | 3.58 | 4.82 | 89.61                | 95.32                | 96.99                | 2.54            | 2.43            | 3.58            | 1.69         |
| CrSI     | 3.75 | 4.81 | 86.02                | 99.71                | 96.96                | 2.75            | 2.45            | 3.75            | 1.20         |

**Table S2.** Calculated anisotropy energy parameter A for the pristine and 1.0 hole-doping CrSX (X=halogen atoms).

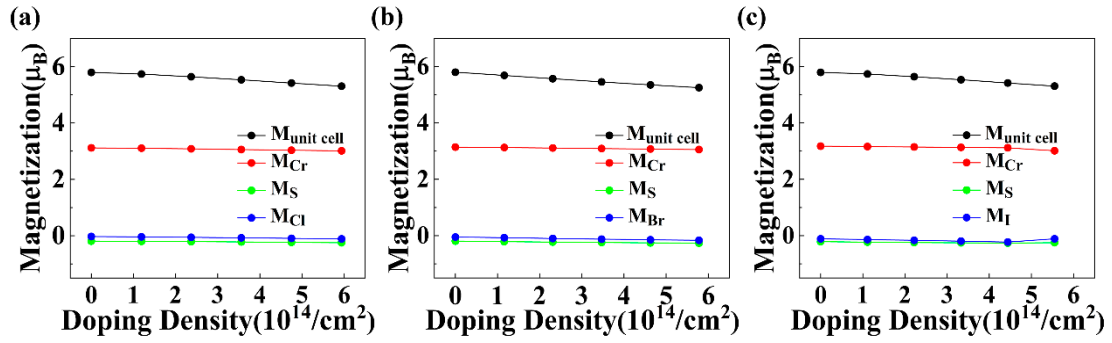
| anisotropy<br>energy<br>parameter | CrSCl    |          | CrSBr    |          | CrSI     |          |
|-----------------------------------|----------|----------|----------|----------|----------|----------|
|                                   | Pristine | 1.0 hole | Pristine | 1.0 hole | Pristine | 1.0 hole |
| A (μeV)                           | 6.69     | 35.44    | 60.98    | 565.64   | 638.51   | 474.62   |

**Table S3.** Calculated in-plane lattice constants, bond lengths, bond angles and band gaps of the critical hole-doped CrSX (X = Cl, Br, I) monolayers.

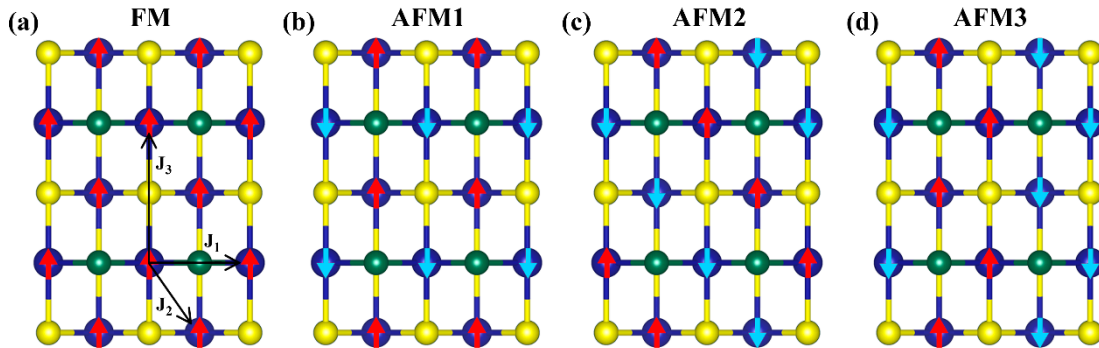
| critical hole-<br>doped | a(Å)  | b(Å)  | $\theta_1(^{\circ})$ | $\theta_2(^{\circ})$ | $\theta_3(^{\circ})$ | $l_1(\text{Å})$ | $l_2(\text{Å})$ | $l_3(\text{Å})$ |
|-------------------------|-------|-------|----------------------|----------------------|----------------------|-----------------|-----------------|-----------------|
| CrSCl<br>(0.05h)        | 3.481 | 4.822 | 93.433               | 92.367               | 97.452               | 2.390           | 2.412           | 3.481           |
| CrSBr<br>(0.007h)       | 3.584 | 4.822 | 89.654               | 95.282               | 97.034               | 2.542           | 2.425           | 3.584           |
| CrSI<br>(0.006h)        | 3.749 | 4.808 | 86.023               | 99.714               | 96.964               | 2.748           | 2.452           | 3.749           |



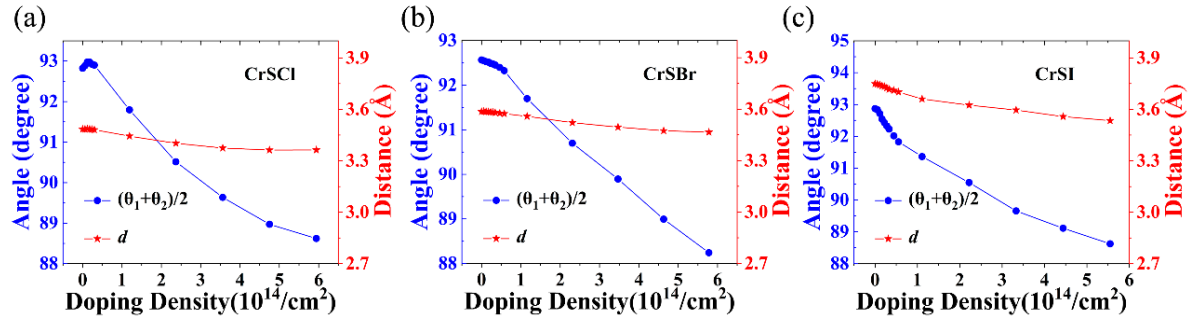
**Figure S1.** The spin polarized charge density for the pristine (a)–(c) CrSX (X = Cl, Br, I) monolayers and 1.0 hole-doped (d) CrSCl, (e) CrSBr, and (f) CrSI monolayers, respectively. The spin-up density is shown in yellow and the spin-down density is shown in blue, respectively. The iso-surface value is set to  $0.005 \text{ e}\text{\AA}^{-3}$ .



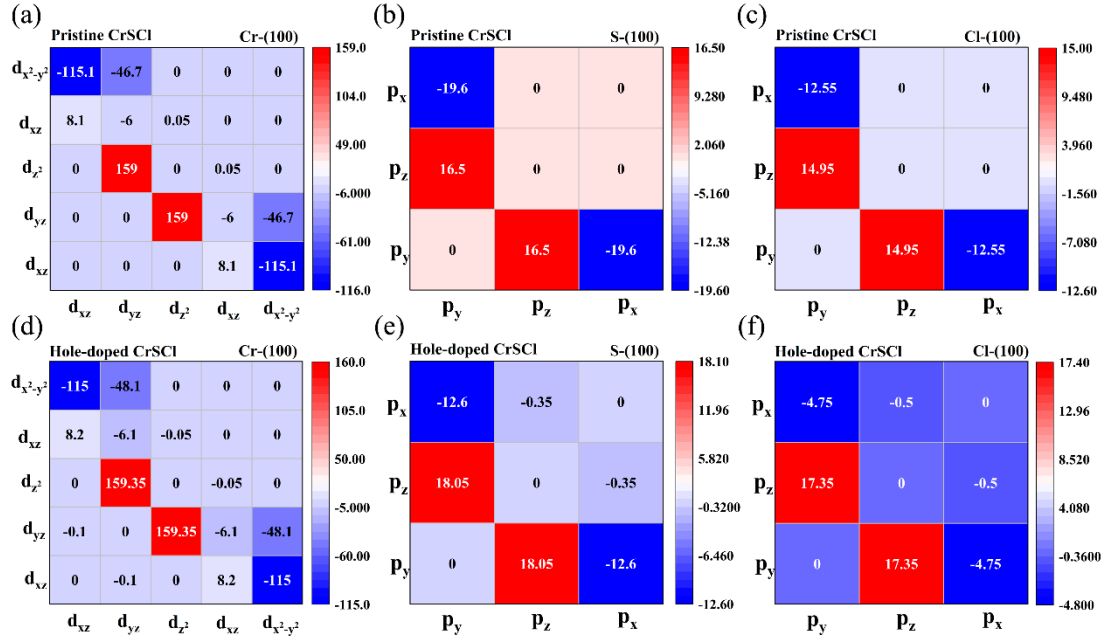
**Figure S2.** Magnetic moment as a function of the hole-doping density for 2D (a) CrSCl, (b) CrSBr, and (c) CrSI.



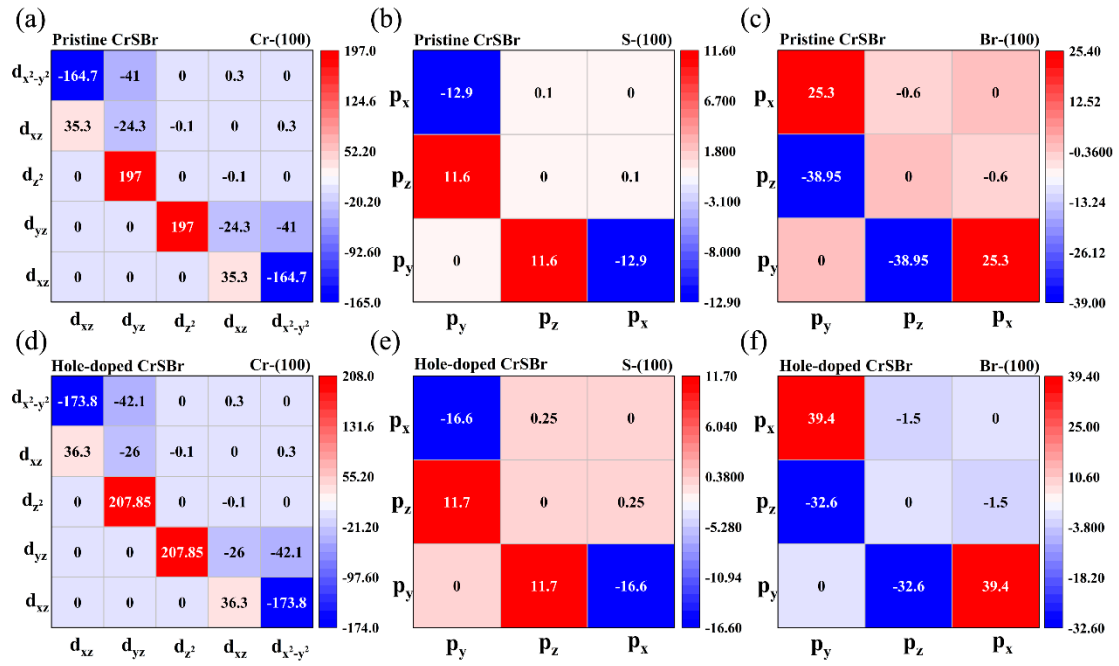
**Figure S3.** Schematic illustrations of four feasible types of magnetic configurations are considered for 2D CrSX (X=Cl, Br, I): (a) FM, (b) AFM1, and (c) AFM2, and (d) AFM3. Red and blue arrows denote spin up and spin down directions, respectively.



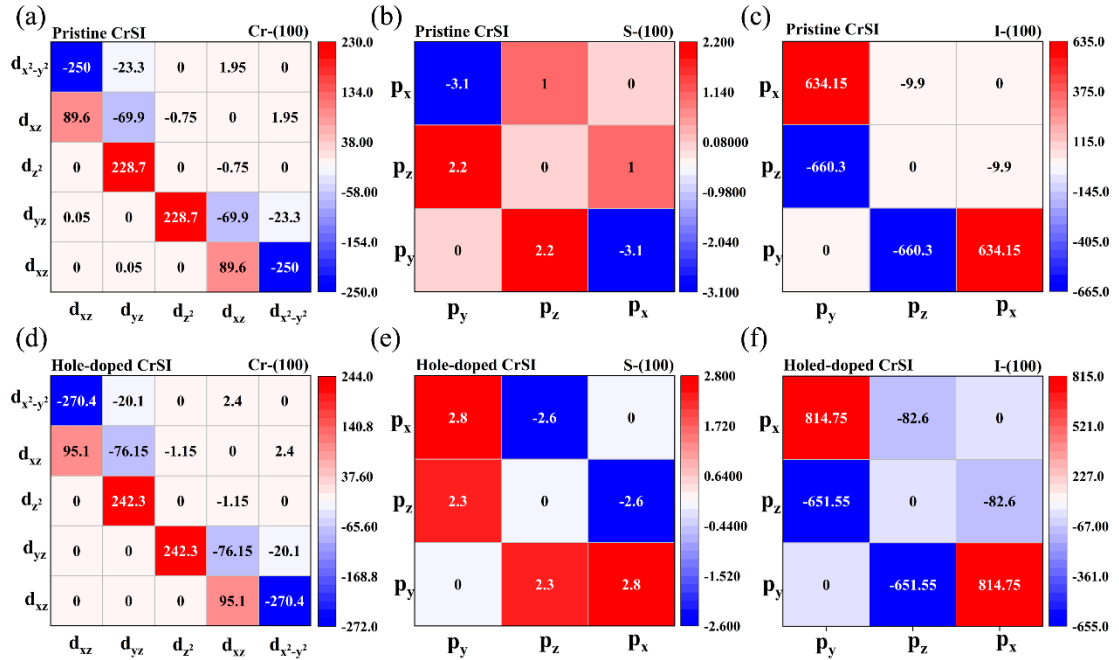
**Figure S4.** Hole-doping dependence of the nearest Cr–Cr distance and the average bond angle of cation-anion-cation for the 2D (a) CrSCl, (b) CrSBr, and (c) CrSI monolayer.



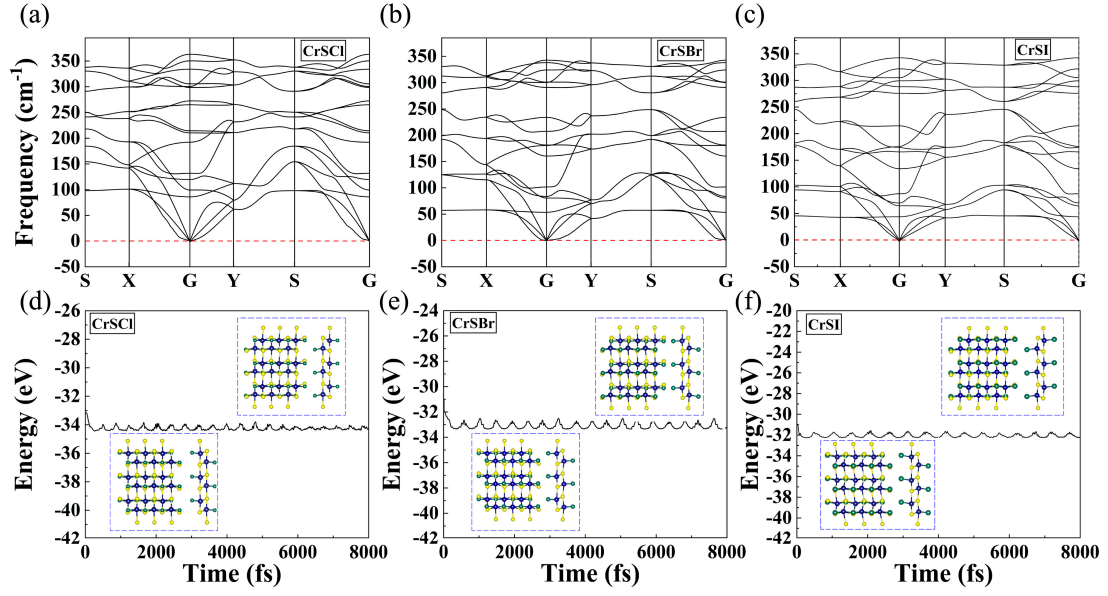
**Figure S5.** Orbital-projected contribution to MAE for (a) Cr, (b) S, and (c) Cl atom for the pristine CrSCl monolayer with spin orientation along (100) direction. Orbital-projected contribution to MAE for (d) Cr, (e) S, and (f) Cl atom for the hole-doped CrSCl monolayer with spin orientation along (100) direction.



**Figure S6.** Orbital-projected contribution to MAE for (a) Cr, (b) S, and (c) Br atom for the pristine CrSBr monolayer with spin orientation along (100) direction. Orbital-projected contribution to MAE for (d) Cr, (e) S, and (f) Br atom for the hole-doped CrSBr monolayer with spin orientation along (100) direction.



**Figure S7.** Orbital-projected contribution to MAE for (a) Cr, (b) S, and (c) I atom for the pristine CrSI monolayer with spin orientation along (100) direction. Orbital-projected contribution to MAE for (d) Cr, (e) S, and (f) I atom for the hole-doped CrSI monolayer with spin orientation along (100) direction.



**Figure S8.** Phonon spectra of hole-doped 2D (a) CrSCl, (b) CrSBr, and (c) CrSI. AIMD simulations of hole-doped 2D (d) CrSCl, (e) CrSBr, and (f) CrSI at 300 K.