Supplementary Material

Two-Dimensional Hexagonal Chromium Chalco-halide with Large Vertical Piezoelectricity, High-temperature Ferromagnetism and High Magnetic Anisotropy

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Contents：

Method to calculate carrier mobility:

The carrier mobility of 2D materials can be estimated by the relation as shown below:[1]

$$\mu = \frac{2e\hbar^3 C}{3k_{\mathrm{B}}T\left|m^*\right|^2 E_1^2},$$

where e is the electron charge, $\hbar$ is the reduced Planck constant, C is the in-plane elastic modulus, K_{B} is the Boltzmann constant, and T is the temperature. The temperature T is set to 300 K in this work. Moreover, m^* is the carrier effective mass. E_1 is the deformation potential proposed by Bardeen and Shockley [2], which denotes the shift of the band edges induced by a small strain. Assuming that the electronic structure is in parabolic form, we can deduce the carrier effective mass m^* from the expression: $m^* = \hbar^2 / \left(\partial^2 E(k) / \partial k^2\right)$, where $E(k)$ is the energy and k is momentum. The effective mass of hole (m_h^*) and electron (m_e^*) can be obtained by conducting a quadratic fitting of the energy band curvature around VBM and CBM, respectively.

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The changes of VBM and CBM positions with respect to vacuum level are presented in Figure S2. The slopes of the valence edge and conduction band edge are the deformation potentials of electron (E_{1e}) and hole (E_{1h}) states, respectively.

Table SI. Calculated deformation potential constants E_{1e} and E_{1h}, effective mass m_h* and m_e*, carrier mobility μ_e and μ_h at 300 K. Subscripts "e" and "h" denote "electron" and "hole", respectively. The units are eV, m_e, and $cm^2 V^{-1} s^{-1}$, respectively.

Structure	E_{1e}	E_{1h}	m_e* (K←Γ→M)	m_h* (K←Γ→M)	μ_e (K←Γ→M)	μ_h (K←Γ→M)
CrSeBr	8.55	4.39	0.981/0.981	0.367/0.367	10.15/10.15	275.2/275.2
CrTeI	11.42	4.03	1.395/1.395	0.491/0.524	2.24/2.24	145.2/127.5
MoS$_2$	11.14	5.61	0.481	0.60	62.5	158.4
MoS$_2$[3]	11.12	5.53	0.475	0.585	66.24	176.19

Figure S1:

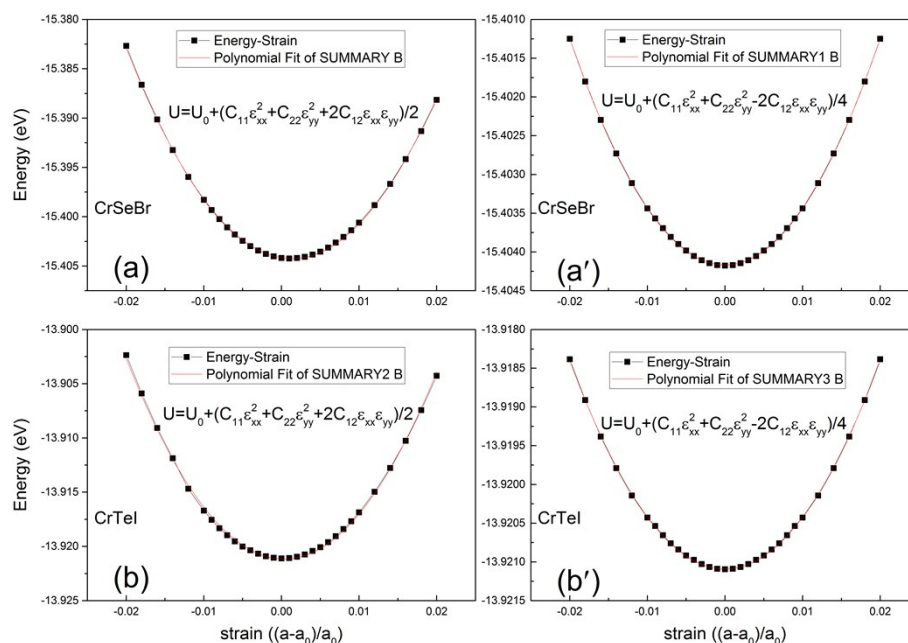**Figure S1.** The energy-vs.-strain curves and theirs fitting under various strains for (a, a′) CrSeBr and (b, b′) CrTeI monolayers.
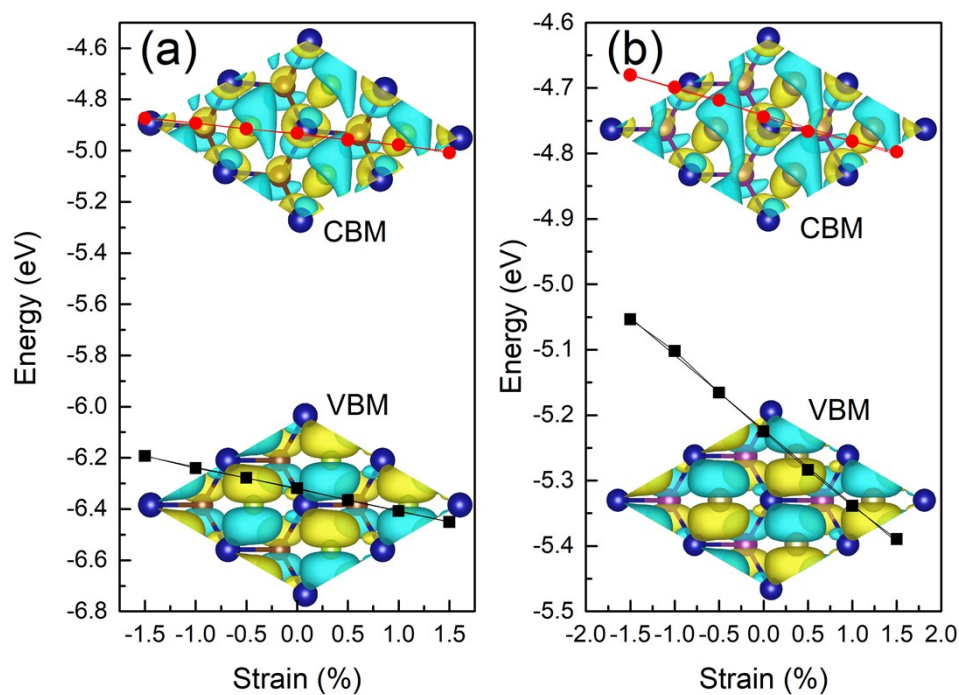
Figure S2:

Figure S2. Linear fitting of deformation potential for (a) CrSeBr and (b) CrTeI monolayer, respectively. The VBM and CBM of monolayers align with respect to the vacuum energy as a function of strain $\Delta a/a$. To reduce computational cost, GGA+U functional with inclusion of the SOC were adopted to calculate the deformation potential constant. The insets are plots of the real-space wave function of VBM and CBM of monolayers. Obviously, the VBM are mainly composed of p_{xy} orbitals of anions, and CBM are made up of p_z orbitals of anions and Cr-$3d_z^2$.
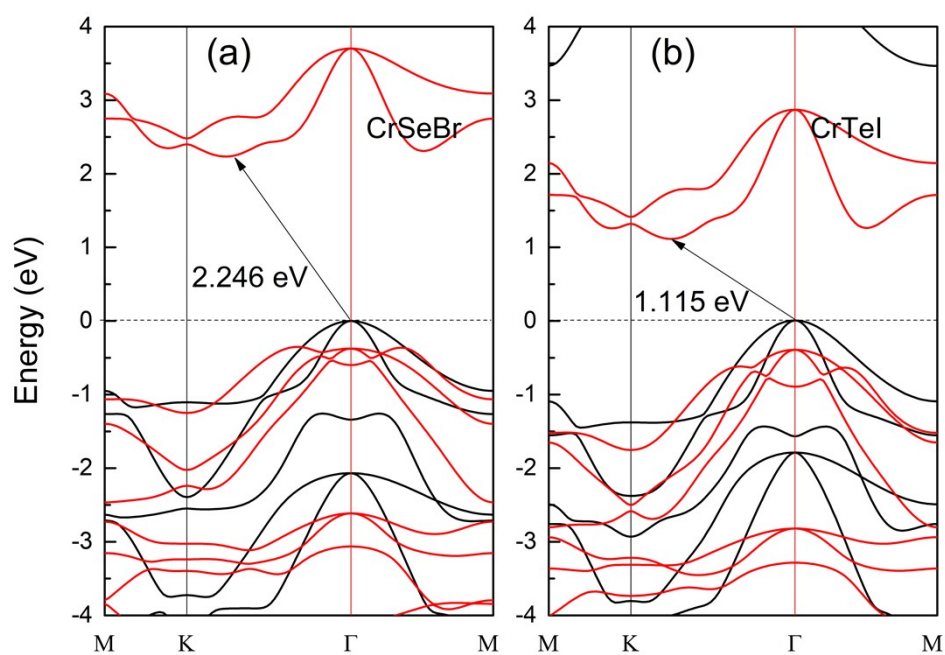
Figure S3:

Figure S3. Calculated electronic band structures for (a) CrSeBr and (b) CrTeI monolayers. GGA+*U*+HSE06 functional without SOC were adopted for calculations. The red and black lines correspond to up-spin and down-spin channels, respectively.
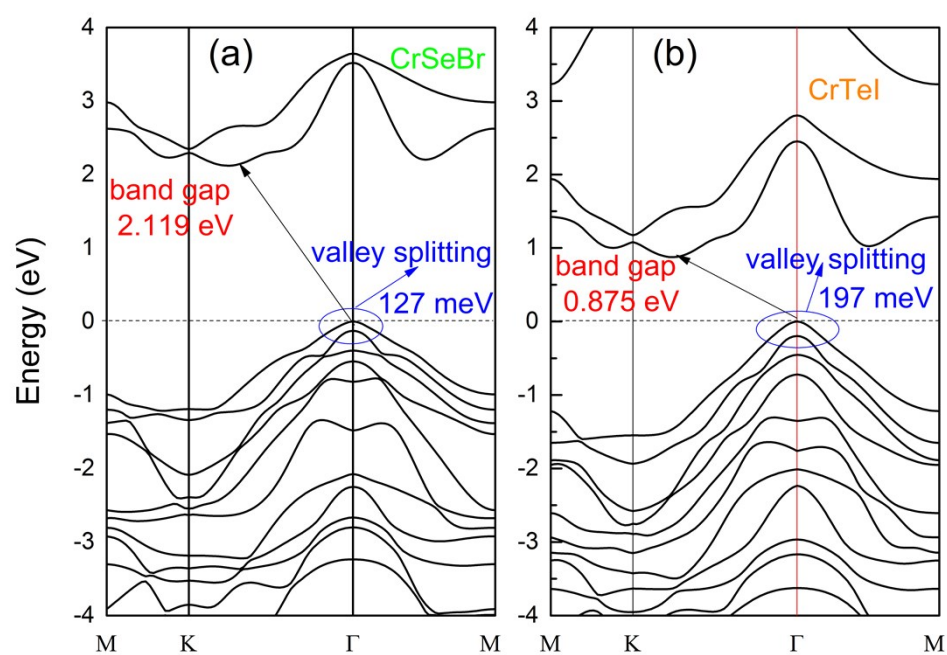
Figure S4:

Figure S4. Calculated electronic band structures for (a) CrSeBr and (b) CrTeI monolayers. GGA+U+HSE06 functional with SOC were adopted for calculations. The red and black lines correspond to up- and down-channels, respectively.
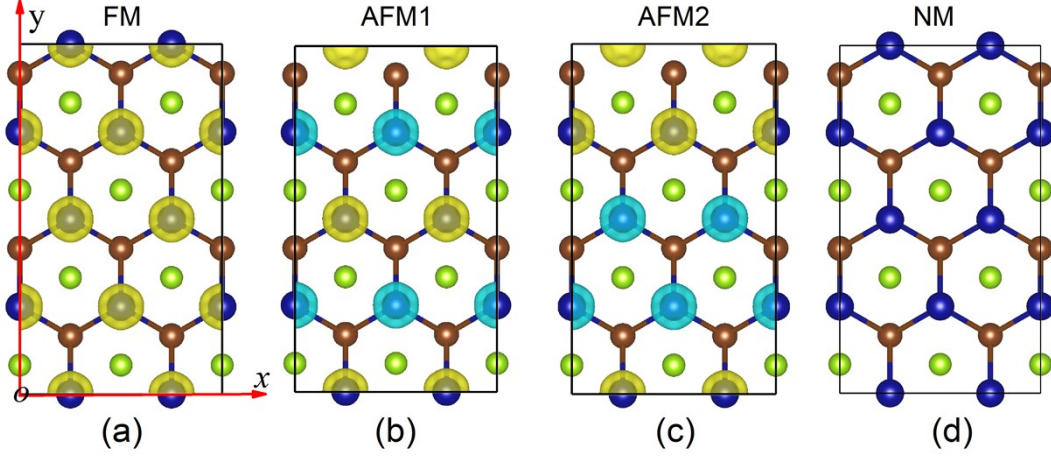
Figure S5:

Figure S5. Schematic representation of four magnetic order: (a) ferromagnetic (FM), (b) anti-ferromagnetic 1 (AFM1), (c) AFM2, and (d) nonmagnetic states, respectively, used for calculation of exchange parameters J_i. The blue and yellow regions respectively corresponds the down- and up-spin polarized densities isosurface with a value of 0.05 e/Bohr^3.

Method to calculate exchange coupling parameter J_i:

If only nearest-neighbour (NN) and next-nearest-neighbour (NNN) exchange interactions are taken into account, the Hamiltonian is written as

$$H = H_0 + \sum_{i,j} J_1 M_i M_j + \sum_{i,k} J_2 M_i M_k$$

According to the magnetic configurations shown in Figure S1 and the expression of Hamiltonian, the total energies of these magnetic configurations are expressed as:

$$\begin{cases} E_{FM} = E_0 + S^2(6J_1 + 6J_2) \\ E_{AFM1} = E_0 + S^2(-2J_1 - 2J_2) \\ E_{AFM2} = E_0 + S^2(2J_1 - 2J_2) \end{cases}$$

where S represents the unpaired spin number on each Cr atom (S=1.5 in our calculations). E_{FM}, E_{AFM1}, E_{AFM2}, and E_{NM} are the energies of the FM, AFM1, AFM2, and nonmagnetic states, respectively. The calculation of the coupling parameter J

used the GGA+U method. When nearest-neighbour and next-nearest-neighbour interactions are only taken into account, the exchange parameters J_1 and J_2 to be:

$$\begin{cases} J_1 = (E_{AFM2} - E_{AFM1})/4S^2 \\ J_2 = (E_{FM} + E_{AFM1} - 2E_{AFM2})/8S^2 \end{cases}$$
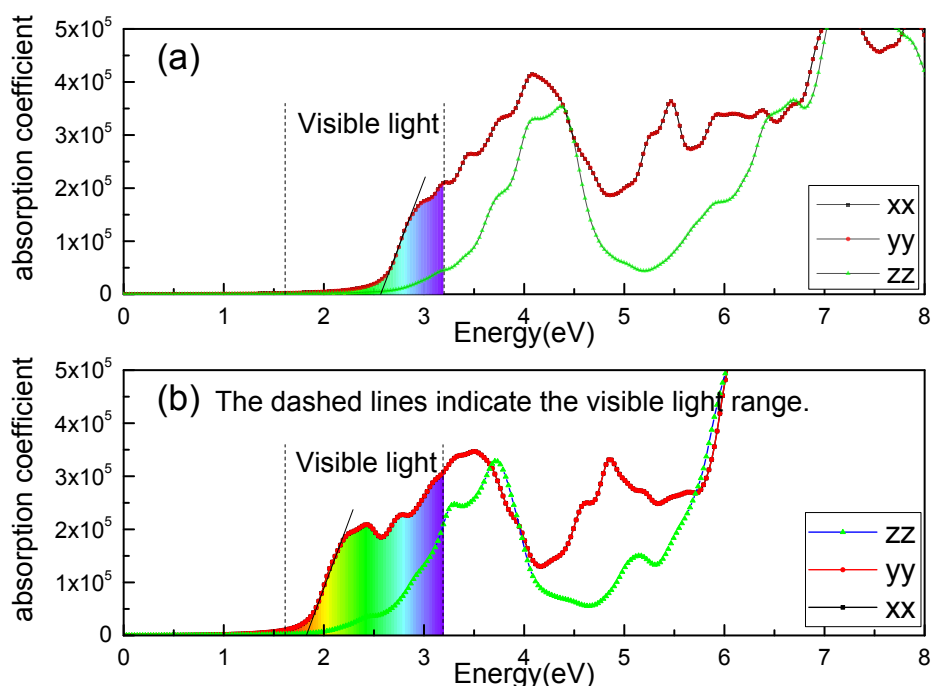
Figure S6:

Figure S6. Calculated absorption spectra along different directions for (a) CrSeBr and (b) CrTeI monolayer, respectively. GGA+U+HSE06 functional without SOC were adopted to calculate the absorption spectra. According to the AM 1.5 solar spectrum [4], almost all the contribution of solar power reaching the earth's surface comes from the photons below 3.4 eV. Apparently, the optical absorption of CrTeI covers the entire visible light spectrum, indicating that CrTeI can be a promising candidate for solar-harvesting materials.

[1] Xi J, Long M, Tang L, et al. First-principles prediction of charge mobility in carbon and organic nanomaterials[J]. Nanoscale, 2012, 4(15): 4348-4369.

[2] Bardeen J, Shockley W. Deformation potentials and mobilities in non-polar crystals[J]. Physical review, 1950, 80(1): 72.

[3] Cai, Y.; Zhang, G.; Zhang, Y. W. Polarity-reversed robust carrier mobility in monolayer MoS_2 nanoribbons. J. Am. Chem. Soc. 2014, 136(17), 6269-6275.

[4] The AM, 1.5G, spectrum was extracted from National Renewable Energy Laboratory website: http://rredc.nrel. gov/solar/spectra/am1.5.