Supplemental Material for

Electric control of Chern number in valley-polarized quantum anomalous Hall insulators

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Supplementary Note 1: The underlying physical mechanism for spontaneous valley polarization in ferromagnetic semiconductors.

Due to the spontaneous valley polarization is induced by SOC with the intra-atomic interaction $\hat{L}\bullet\hat{S}$, we thus consider SOC as a perturbation, which can be written as $\hat{H}^0_{SOC}+\hat{H}^1_{SOC}=\lambda\hat{L}\bullet\hat{S}$.[1, 2] Here, $\hat{L}$ and $\hat{S}$ are orbital angular moment and spin angular moment, respectively.

$$\hat{H}^0_{SOC}=\lambda\hat{S}_{z'}(\hat{L}_z\cos\theta+\frac{1}{2}\hat{L}_+e^{-i\phi}\sin\theta+\frac{1}{2}\hat{L}_-e^{+i\phi}\sin\theta),$$

$$\hat{H}^1_{SOC}=\frac{\lambda}{2}(\hat{S}_{+'}+\hat{S}_{-'})(-\hat{L}_z\sin\theta+\frac{1}{2}\hat{L}_+e^{-i\phi}\cos\theta+\frac{1}{2}\hat{L}_-e^{+i\phi}\cos\theta)$$

$\hat{H}^0_{SOC}$ is the Hamiltonian that describes the interaction between the same spin states, while $\hat{H}^1_{SOC}$ indicates the interaction between opposite spin states. Given that the K (-K) valleys in valence bands and conduction bands hold identical spin near Fermi level, the effective Hamiltonian of SOC can be written as:

$$\hat{H}_{SOC}=\lambda\hat{S}_{z'}(\hat{L}_z\cos\theta+\frac{1}{2}\hat{L}_+e^{-i\emptyset}\sin\theta+\frac{1}{2}\hat{L}_-e^{+i\emptyset}\sin\theta)$$

where θ and $\emptyset$ are the spin orientations. When the magnetization orientation is perpendicular to the plane ($\theta = 0°$), the Hamiltonian can be simplified as: $\hat{H}_{SOC} = \lambda \hat{S}_z \hat{L}_z$. Due to the orbital composition of K (-K) valleys in vicinity of Fermi level is composed of $d_{x2-y2/xy}$ of Cr atoms We thus adopt

$$|\phi_{c/v}^{\tau}\rangle = \sqrt{\frac{1}{2}}\left(|d_{x^2-y^2}\rangle + i\tau|d_{xy}\rangle\right)$$

as orbital basis, where $\tau = \pm 1$ represent valley index, the subscript $c(v)$ indicates conduction (valence) band. Under the influence of SOC, the energy level of K/K' valleys in conduction and valence band can be described as follows:

$$E_c^{\tau} = \langle \phi_c^{\tau}|\hat{H}_{soc}|\phi_c^{\tau}\rangle,\ E_v^{\tau} = \langle \phi_v^{\tau}|\hat{H}_{soc}|\phi_v^{\tau}\rangle$$

It is well established that $\hat{L}_z|d_{x^2-y^2}\rangle = 2i|d_{xy}\rangle,\ \hat{L}_z|d_{xy}\rangle = -2i|d_{x^2-y^2}\rangle$. And the valley polarization of CBM and VBM are $E_{c/v}^{K} - E_{c/v}^{-K} = i\langle d_{x^2-y^2}|\hat{H}_{SOC}|d_{xy}\rangle - i\langle d_{xy}|\hat{H}_{SOC}|d_{x^2-y^2}\rangle = 4\alpha$, $\alpha = \lambda\langle d_{x^2-y^2}|\hat{S}_z|d_{x^2-y^2}\rangle$. Hence, the valley polarization can occur in both CBM and VBM. Here, we observe that a difference in the values of valley polarization between the CBM and VBM, which mainly arises from the warped band structure caused by band inversion at K valley.

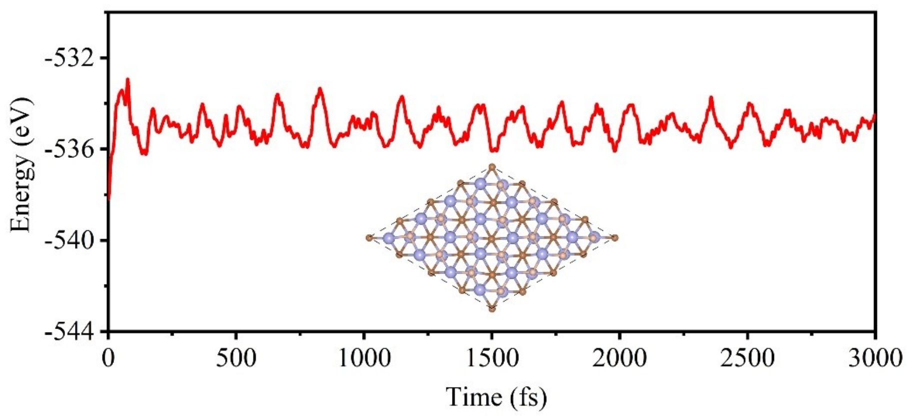

FIG. S1. Fluctuations of the total energy of Cr_2COH monolayer at 300 K. Inset is the snapshot taken from the end of the MD simulation.

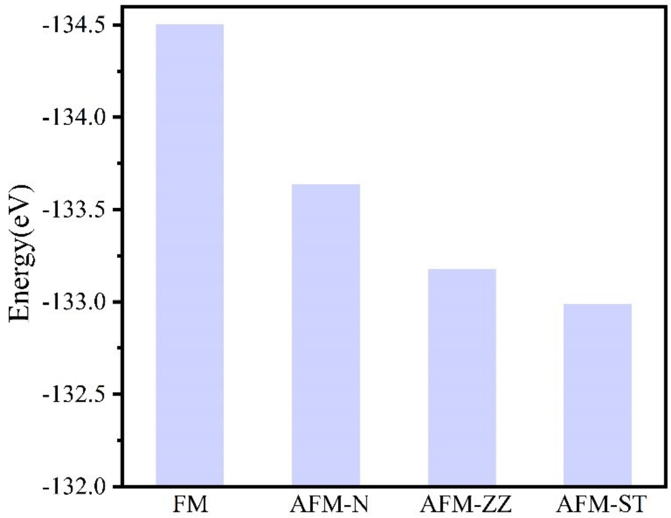

FIG. S2. Total energies of FM and various AFM configurations for the Cr_2COH monolayer.

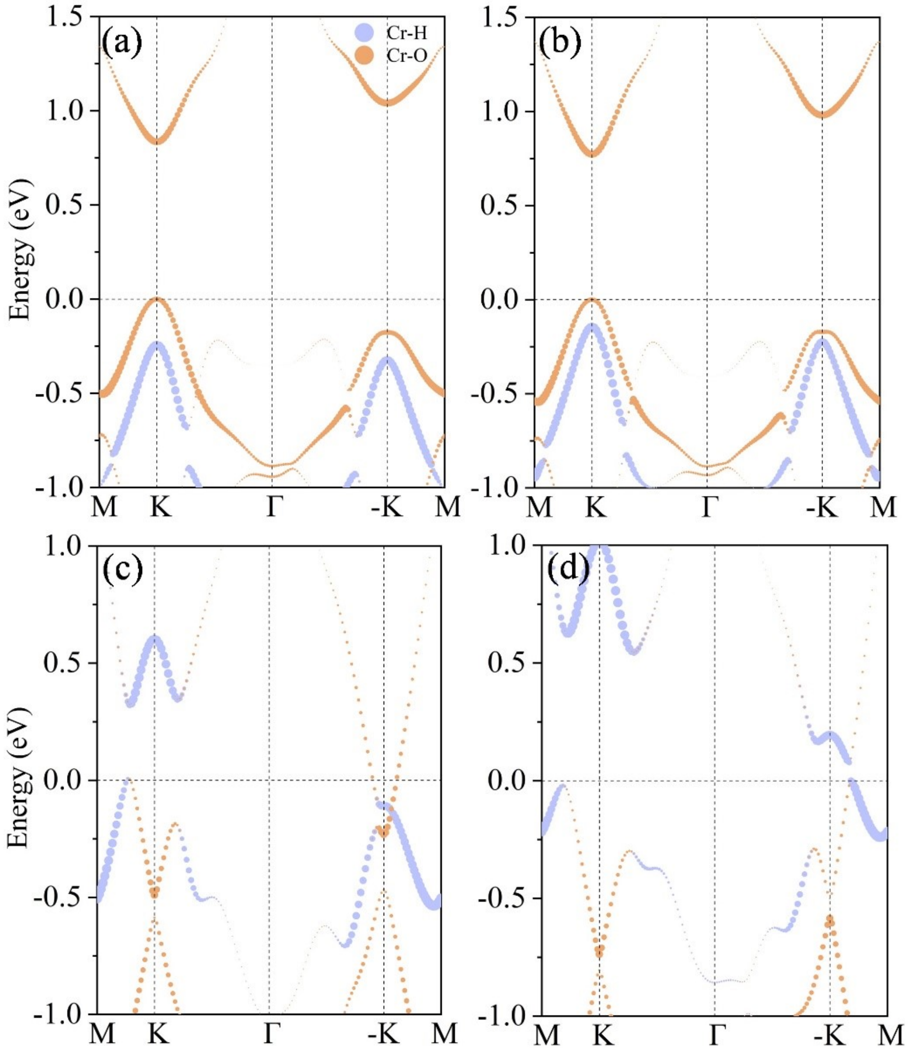

FIG. S3. The *d*-orbitals-project band structure of Cr_2COH monolayer under electric field of (a) -0.2 V/Å, (b) -0.15 V/Å, (c) 0.15 V/Å and (d) 0.2 V/Å.

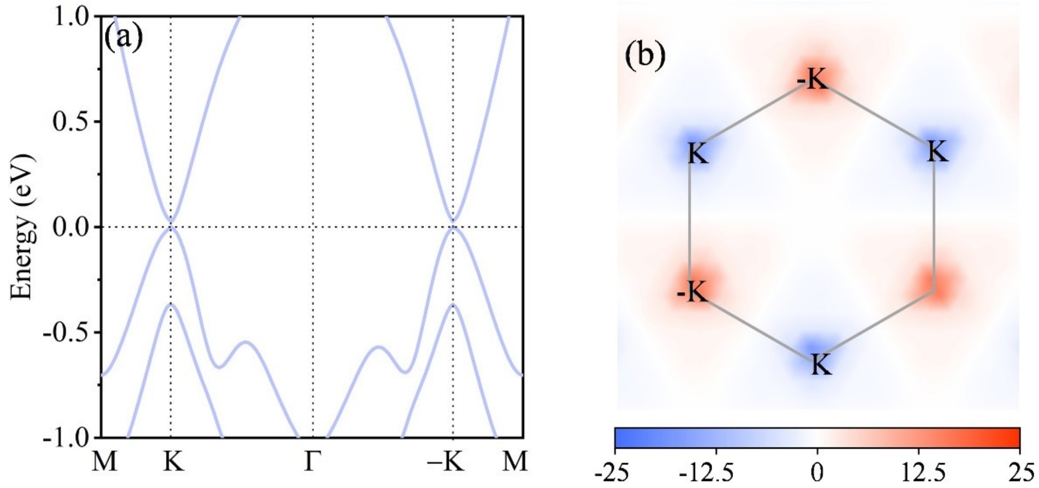

FIG. S4. (a) Band structures and Berry curvature of Cr_2COH monolayer with in-plane magnetization orientation when considering SOC.

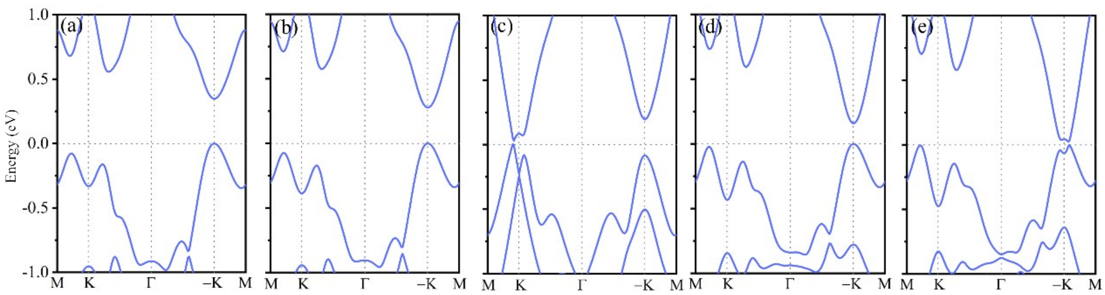

FIG. S5. Band structures with considering SOC under biaxial strain of (a) -2%, (b) -1%, (c) 0%, (d)1% and (e) 2%.

1. Dai, D.; Xiang, H.; and Whangbo, M. H. J. Comput. Chem. **29**, 2187 (2008).
2. Whang, M.; Gordon, E.; Xiang, H.; Koo, H.; Lee, C. Acc. Chem. Res. **48**, 3080 (2015).