Supporting Information for

Ferroelectricity and Large Rashba Splitting in Two-Dimensional Tellurium

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SI Note I:

For Abm2 space group of FE phase, the corresponding point group symmetry is C_{2V} here, which includes 2-fold rotation symmetry of the z-axis and mirror symmetry of [110]-direction. This mirror symmetry constrains the direction of polarization field, which directly leads to the spin polarization with only S_z component, according to the Rashba toy model below:

$$H \propto \left(\vec{E} \times \boldsymbol{k}\right) \cdot \sigma,$$

where $\vec{E}$ and σ represent the electric filed and spin Pauli matrix, respectively. As discussed in the main text, FE phase possess the in-plane polarization field and thus the above Hamiltonian can be written as $\{\sigma_z(-E_x, k_y) - \sigma_y(-E_x, k_z)\}$. For simplicity, we assume that the polarization field is parallel to x'-axis and thus E_x, terms are left in the Hamiltonian. For 2D materials, the k_z momentum is meaningless and thus the system will only keep S_z component, i.e., $\sigma_z(-E_x, k_y)$. Therefore, near CBM, the spin Hall conductivity can be reasonably calculated according to Eq. (3) in the main text. More interestingly, the spin can be reversed when the direction of E_x, (polarization

field) is reversed through the FE phase transition. Such analyses based on the Rashba toy model capture the key characteristics of the DFT results in Figs. 5 and S7.

SI Note II:

Here we will simply discuss the Dresselhauss and Zeeman-type splitting in our FE system. For 2D system, the Dresselhauss contribution normally writes as:

$$H \propto k_x\sigma_x - k_y\sigma_y,$$

from which we will find in-plane spin components (S_x/S_y). But for the FE phase here, the system has only in-plane polarization and S_x/S_y are found to be zero (Fig. S8) near CBM. Thus, we rationally exclude the Dresselhauss contribution near CBM.

For Zeeman-type splitting, the spin-degenerate pair will split mostly in energy not in *k*-momentum and each band carries the opposite spin component. As shown in Figs. 5 and S8, near band edge, the key feature of FE system is the *k*-momentum band-splitting, quite different from the Zeeman-type bands. Therefore, we also exclude the Zeeman-type contribution near CBM.

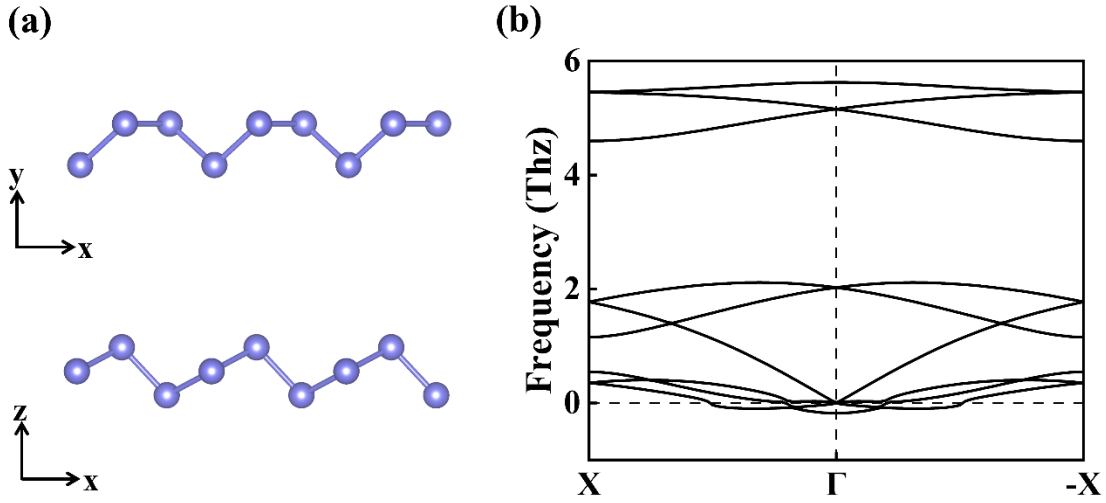

Fig. S1. (a) Top view and side view of 1D α-Te. (b) The phonon spectrum of 1D α-Te with one imaginary chiral-rotation mode at Γ.

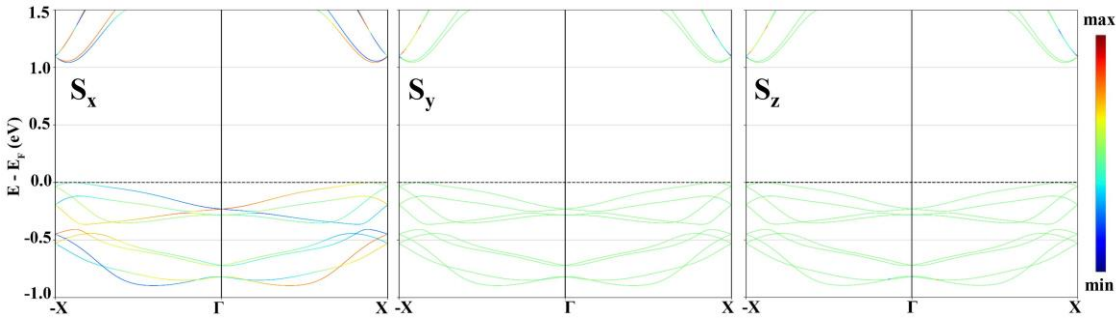

Fig. S2. Spin-resolved bands of 1D s-Te.

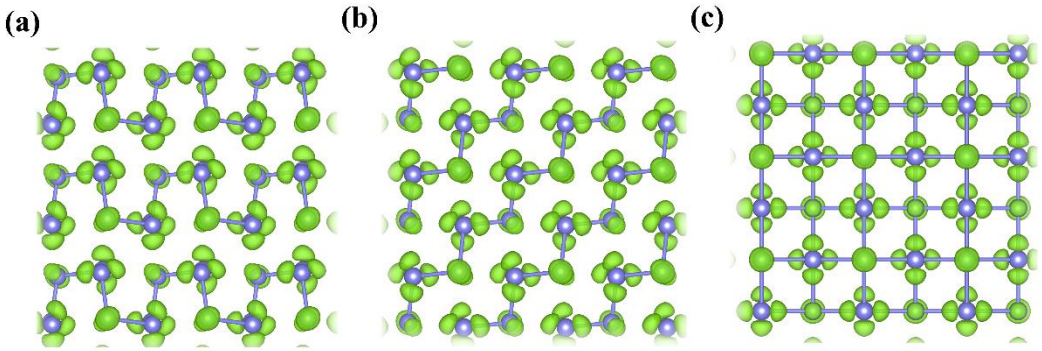

Fig. S3. ELF distribution of (a) phase A, (b) phase C and (c) phase B. The isosurface is 0.9 e/Bhor^3.

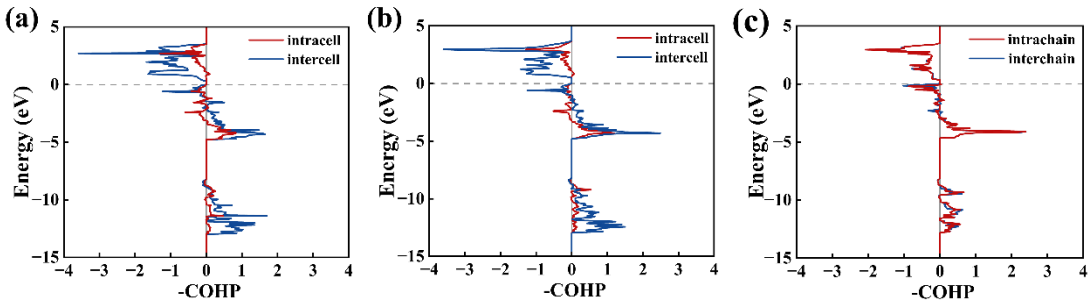

Fig. S4. COHP results. (a)−(b) intra/inter-cell interactions for phase A and C, respectively. (c) Intra/inter-chain interactions for the phase B.

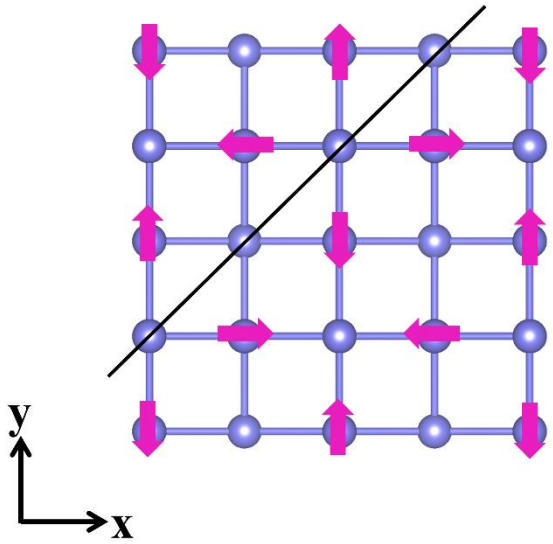

Fig. S5. The schematic figure of vibration mode λ_4. The black line denotes the remaining Te-chain after the distortion. Such structure is unstable during optimization, and it will transition back to phase B.

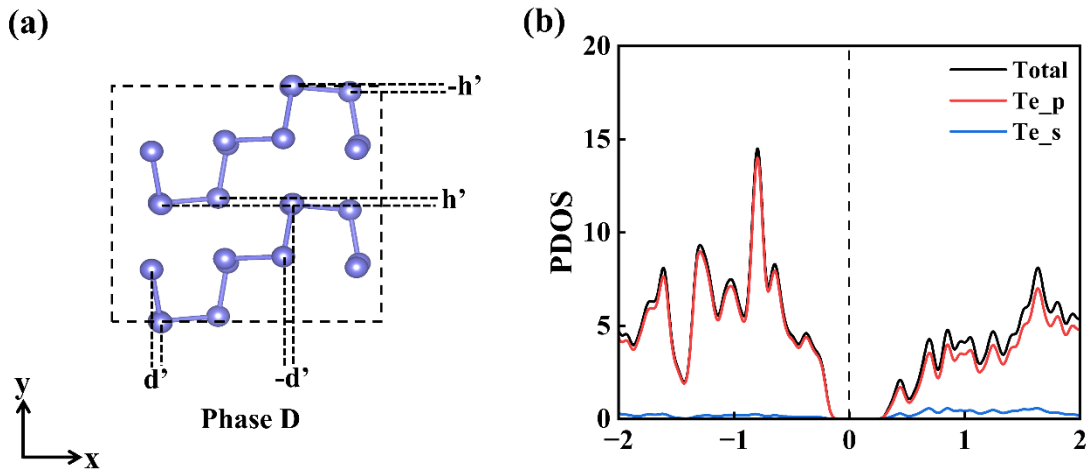

Fig. S6. (a) Top view of phase D in a 2 × 1 × 1 supercell. (b) Partial density of states for phase D, exhibiting the semiconducting behavior.

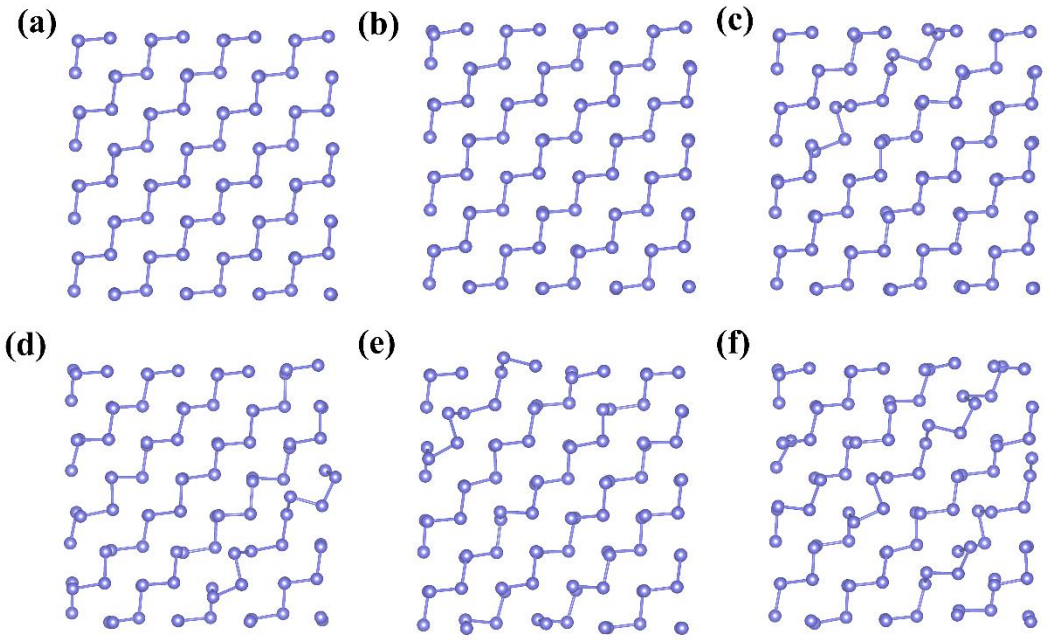

Fig. S7. AIMD results of phase C structure at different temperatures: (a) 20 K, (b) 30 K, (c) 40 K, (d) 50 K, (e) 60 K and (f) 70 K.

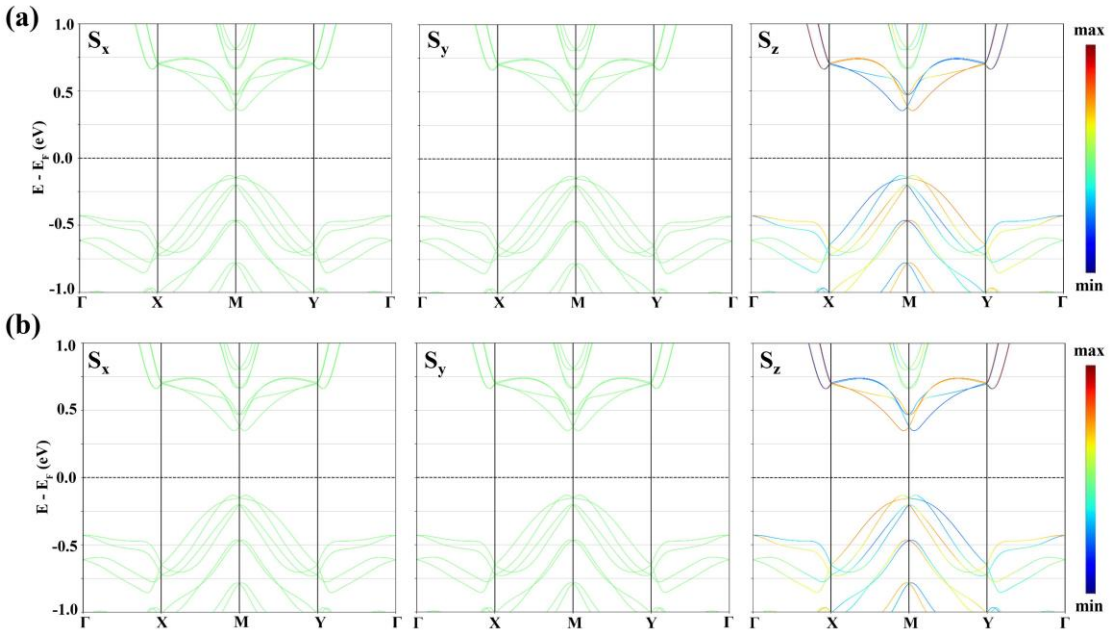

Fig. S8. (a)−(b) Band structures of phase C and C', respectively. The spin components are projected. S_x and S_y components are found to be zero. It is obvious to see that the spin polarization is reversed for phase C and C', according well with the toy model.

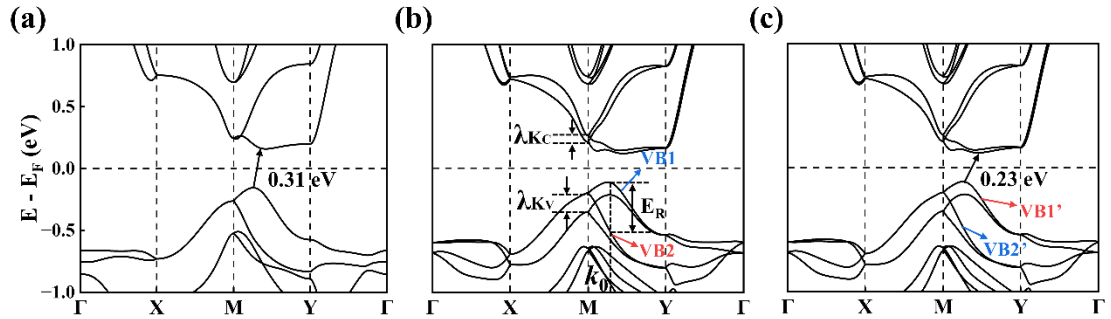

Fig. S9. (a) Band structure of A/A' phases without SOC. (b)−(c) Band structures of A/A' phases with SOC, respectively. Under phase transition from A to A', the spin polarization reverses near VBM and CBM.