electronic supplementary information for

Two-dimensional ferroelectricity and switchable spin-textures in ultra-thin elemental Te multilayers†

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**Fig. S1** Side view of the three-layer α, β, γ, δ, and ε phases of the (a-e) XZ plane and (f-j) YZ plane. (k-o) Top view of the corresponding phases of the XY plane.
Fig. S2 COHP for intra- and interchain Te bonds in the bulk Te, bilayer α phase and bilayer β phase.
Note1: The details for calculation of 2D polarization value.

To obtained 2D polarization value, we defined it as the number of electric dipoles (C*m) per square meters (C/m) with corresponding to 3D value (the number of electric dipoles(C*m) per cubic meters (C/m^2)). We can link these two quantity with an effective thickness. The polarization direction is defined from positive charge to negative one.

**Fig. S3** Layer-resolved polarization value of the Te α phase with different layer numbers. Different vdW methods were adopted to examine the variations.
**Fig. S4** Phonon spectra for the (a-b) 3L, (c-d) 4L, (e-f) 5L, (g-h) 6L, (i-j) 7L Te α, β phases and the (k-l) bulk Te phase.
**Fig. S5** Schematic drawing of energy surface for the highest valence band in bilayer Te α phase.
**Fig. S6** (a-b) Snapshots (side view) of the molecular dynamic simulations at 300 K and 600 K. (c-d) Energy and temperature fluctuation with respect to the time for the bilayer α phase at 300 K and 600 K.
Fig. S7 (a) Illustration of MD simulation configuration for the bilayer $\alpha$ phase at 0K. Averaged distance value is linear related to polarization value. (b) Temperature dependence of averaged polarization $<P_i>$ of bilayer $\alpha$ phase. Here, $<P_i>$ at different temperatures has been normalized with respect to $<P_i>_{T=0K} = P_S$. The points above 650K means structural collapse resulting from temperature beyond melting point.
Note2: Optimized lattice parameters of bulk, $\alpha$ and $\beta$ phase within different vdW methods

To find the most accurate vdW method descriptions, we adopted Tkatchenko-Scheffler (TS) vdW method, vdW-DF level with the optB88 exchange function (optB88-vdW), DFT-D2 method and Many-body dispersion energy method (MBD) to check the geometry results. We see no significant discrepancy with all the vdW interactions and DFT-TS method agree better with experiments data.

**Fig. S8** (a-b) Lattice constants (a and b) of multilayer and bulk Te within different vdW interactions for $\alpha$ and $\beta$ phase
Fig. S9 (a-b) Surface (s) and inner (i) interlayer distances, Δa as marked in Fig. S1a, of multilayer Te for α and (c-d) β phase.
Fig. S10 (a) The layer-dependent formation energy of Te α and β phase within different vdW methods. (b) The layer-dependent barrier defined as the energy difference between β and α phase within different vdW methods.