## Supporting Information

## Dimensionality-Driven Phase Engineering in 2D Noble Metal

Chalcogenides: New Phase via Confined Chemical

## Transformation

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**Figure S1**. (a) EDS of the as-synthesized  $Ag_{4.53}Te_3$ . The inset is the TEM image. (b) The HRTEM sample. (c) The complete HRTEM image and the corresponding selected area electron diffraction patterns.



**Figure S2**. The top view of the Te frame in  $Ag_{4.53}Te_3 2x2x2$  unit cell. In order to observe the Te frame clearly, we did not draw Ag atoms in the lattice structure.



Figure S3. The anion frame change in the perspective projected in [0001] direction.



Figure S4. The side and top view of the atomic structure of Ag<sub>4.53</sub>Te<sub>3</sub> 2x2x2 unit cell.



Figure S5. The contact potential difference tested using KPFM. (a) The sample nanoflake used for testing, with blue boxes indicating the testing range. (b) Surface potential distribution of the sample. (c) Contact potential difference of the sample.

We employed Kelvin probe force microscopy (KPFM) to measure the contact potential difference (CPD) of the synthesized  $Ag_{4.53}Te_3$ . Based on the CPD measurements and the relevant equation,

$$V_{CPD} = \frac{\phi_{\rm tip} - \phi_{\rm sample}}{-q}$$

the work function of  $Ag_{4.53}Te_3$  was calculated to be approximately 5.9 eV.



Figure S6. Transfer curve of a typical long-channel 2D Ag<sub>4.53</sub>Te<sub>3</sub> transistor

We chose Cr/Au (25/75 nm) for the metal contacts because Cr has a relatively high workfunction that can reduce the contact resistance in p-type transistors.



Figure S7. Schematic diagram of piezoelectric direction



Figure S8. Standard contact PFM investigation for 2D  $Ag_{4.53}Te_3$ .



Figure S9. Average amplitude variations versus applied voltages.

The amplitude images captured at varying tip voltages, along with their corresponding topography and phase images, are presented in **Fig. S5**. Based on **Fig. S6**, the vertical piezoelectric coefficient ( $d_{eff}$ ) is approximately 28.2 pm·V<sup>-1</sup>. We have compared some 2D materials, as shown in the table S1. The results indicate that Ag<sub>4.53</sub>Te<sub>3</sub> exhibits remarkable piezoelectricity, which is superior to that of other related 2D materials.

2D Materials	Piezoelectric
	coefficient d <sub>33</sub>
Multilayered	14.76 pm·V <sup>-1</sup>
CuInP <sub>2</sub> S <sub>6</sub> <sup>1</sup>	
2D g-C <sub>3</sub> N <sub>4</sub> <sup>2</sup>	1 pm·V <sup>-1</sup>
2D PbO <sup>3</sup>	30 pm·V <sup>-1</sup>
MoSeTe <sup>4</sup>	6.217 pm·V <sup>-1</sup>
MoTe <sub>2-x</sub> <sup>5</sup>	2.4 pm·V <sup>-1</sup>
MoTe <sub>2</sub> <sup>5</sup>	0.4 pm·V <sup>-1</sup>
SnS <sub>2</sub> <sup>6</sup>	2.2 pm·V <sup>-1</sup>

Table S1 Selected reporting of piezoelectric coefficients of 2D materials



Figure S10. (a) Piezoelectric current after multiple devices are connected in series. (b) The current voltage curve of multiple devices connected in series as a function of strain variation.

Synthesis of 2D tellurene crystals.

In a standard procedure, analytical-grade Na<sub>2</sub>TeO<sub>3</sub> (0.00045 mol) and a specific amount of polyvinylpyrrolidone (PVP) are dissolved in double-distilled water (33 ml) at room temperature under magnetic stirring to create a homogeneous solution. The resultant solution is then poured into a Teflon-lined stainless-steel autoclave, which is subsequently filled with an aqueous ammonia solution (25% by weight) and hydrazine hydrate (80% by weight). The autoclave is sealed and maintained at the reaction temperature for a predetermined duration. Following the reaction, the autoclave is allowed to cool naturally to room temperature. The resulting solid, silver-grey products are precipitated by centrifugation at 5,000 revolutions per minute (r.p.m.) for 5 minutes and washed three times with distilled water to remove any residual ions.



Figure S11. Raman spectra of the tellurene.



Figure S12. HRTEM image of the tellurene.

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