# Supporting Information

The Spontaneous Directional Transformations of the Layers and Chemical Bonds: a Study Combining First-principle Calculations and

## Experiments

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### Synthesis and Characterization



Figure S1 Optical image of template-directed Tenanoflakes. Scale bar, 20 µm.

As evidenced in the optical microscope (OM) image depicted in **Figure S1**, trapezoidal Te nanoflakes have been successfully generated on a silicon wafer substrate encompassed by a 100 nm silicon dioxide layer<sup>1</sup>. This image underlines the distinctive trapezoidal morphology that typifies growing Te nanoflakes as observed under the OM. The discernible differentiation of white or gray nanoflakes on the silicon wafer substrate corresponds to varying thicknesses of Te. It is noteworthy that the PtTe<sub>2</sub> nanoflakes also exhibit various colors under OM (Figure 1c), indicative of different thicknesses that strongly depend on the sacrificial templates' thickness.



Figure S2 Low-magnification TEM image of a PtTe<sub>2</sub> nanoflake.



**Figure S3** Thickness statistics of 2D Te nanoflakes and PtTe<sub>2</sub> nanoflakes synthesized from them.



**Figure S4 a** Optical microscope image of incomplete transformation  $PtTe_2$  sample when the molar ratio of  $[H_2PtCl_4]$ :[Te] is less than 1:3. The samples framed by purple dotted lines are not transformed Te nanoflakes. Scale bar, 70 µm **b** Optical microscope image of incomplete transformation  $PtTe_2$  sample, when the molar ratio of  $[H_2PtCl_4]$ :[Te] is between 1:2 and 1:3. Scale bar, 70 µm.

The transformation process and mechanism from Te templates to  $PtTe_2$  nanoflakes are the focus of our research. Through a series of quantitative experiments, it is found that the synthesis of high-purity and uniform  $PtTe_2$  nanoflakes is closely associated with the molar ratio of the reaction precursors, specifically the molar ratio of  $H_2PtCl_4$  to Te. When the molar ratio of  $[H_2PtCl_4]$ :[Te] is less than 1:3, the transformation of the ultrathin Te template is incomplete, and numerous Te nanoflakes are still observable under the optical microscope (see Figure S4a). Increasing the molar ratio of  $[H_2PtCl_4]$ :[Te] to a range between 1:2 and 1:3 eliminates the observation of unconverted Te nanoflakes in the OM (see Figure S4b), indicating the complete consumption and transformation of Te nanoflakes into PtTe<sub>2</sub>. However, considering the atomic ratio in the product PtTe<sub>2</sub>, it is evident that not all Te atoms are converted into Te<sup>2-</sup> in PtTe<sub>2</sub>. Further increase in the molar ratio of  $[H_2PtCl_4]$ :[Te], that is, higher concentration of chloroplatinic acid in the reaction solution, does not lead to significant changes in the product. This confirms the high stability of the synthesized PtTe<sub>2</sub> nanoflakes, with the Te ions no longer reacting with excess Pt ions in the solution. The influence of the molar ratio of the reaction process and the transformation mechanism of Te nanoflakes into PtTe<sub>2</sub><sup>2-4</sup>.

Figure S5 In situ conversion experiment of Te templates on silicon wafers with markers.

Furthermore, the correlation between the Te templates and the  $PtTe_2$  nanoflakes was corroborated via in situ conversion comparative experiments conducted on silicon wafers that had been appropriately labeled. It is distinctly observable, as illustrated in **Figure S5**, that the geometrical configuration of the nanoflakes remains unaltered prior to and subsequent to the reaction. This experimental verification establishes a fundamental framework, facilitating an enhanced comprehension of the reaction mechanism through its amalgamation with computational analyses.



### **Devices and Transport Properties**

**Figure S6 a** Schematic diagram of the structure of a PtTe<sub>2</sub>-based six-terminals device. Inset, optical micrograph of the device and the orientation of applied magnetic filed. Scale bar, 20

 $\mu$ m **b** The IV characteristic of the device. Inset, the schematic diagram of the test device. **c** Temperature dependence of the resistivity of the PtTe<sub>2</sub> device.

In order to preliminarily explore the transport properties of synthesized  $PtTe_2$ , we fabricated six-terminal devices on Si/SiO<sub>2</sub> substrates. The schematic structure of the devices is illustrated in **Figure S6a**, with a representative OM image of a device constructed on a rectangular sheet of ultrathin  $PtTe_2$  nanoflake shown in the inset of **Figure S6a**<sup>5</sup>. **Figure S6b** displays the near-linear I-V curve of the device at room temperature, indicating an ohmic contact between  $PtTe_2$  and the gold electrode. **Figure S6c** presents the temperature-dependent resistance curve of  $PtTe_2$ , spanning the range from 300 K to 70 K. The resistance decreases with decreasing temperature, demonstrating metallic behavior. Below 70 K, the resistance curve exhibits pronounced upturns at low temperatures, which, according to previous studies, are commonly associated with the manifestation of the topological surface state of the material. Notably, around 260 K, the cooling curve exhibits a distinct inflection point, possibly indicating a phase transition in the material.

Figure S7a illustrates the dependence of magnetoresistance on a vertical magnetic field applied in the [0001] direction, revealing weak antilocalization (WAL) behavior<sup>6, 7</sup>. Similar magnetoresistance behavior is observed in the [2I10] direction (refer to Figure S7b). The WAL effect diminishes as the temperature increases, and it is not observable until above 50 K (see Figure S8). WAL is generally regarded as a characteristic of strong spin-orbit coupling. Figure S7c depicts the variation of magnetoresistance with different angles between the magnetic field and the [0001] direction, while Figure S7d demonstrates the magnetoresistances for magnetic fields applied parallel (B//) and perpendicular (B⊥) to the film plane at 2 K. The difference in magnetoresistances between B// and B⊥ is likely related to the confined vertical dimension of the nanoflake.



Figure S7 a Magnetoresistance at different temperatures with magnetic field applied perpendicular to film, where  $R_0$  is resistance in the absence of magnetic field. **b** Magnetoresistance in [2I10] direction at different temperatures with magnetic field applied perpendicular to film, where  $R_0$  is resistance in the absence of magnetic field. **c** Magnetoresistance under parallel magnetic fields at different angles. **d** Magnetoresistance of a PtTe<sub>2</sub> divice at 2 K for magnetic field along with two different directions.



**Figure S8** Magnetoresistance at temperature 80 K-140 K and direction [0001]. At high temperature, the WAL behavior of magnetoresistance can not be observed, and magnetoresistance is almost a constant with the change of magnetic field.

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	Chemical transformation	CVD/CVT		
Complexity	Simple and easy to implement	Complex operation, high		
		equipment cost		
Applicability	Wide applicability	Wide applicability		
Temperature	Low temperature synthesis	High temperature demand		
demand				
Controllability	Limited to solvent selection	Strong controllability		
Purity	Relatively low purity	High purity		
Material size	Small material size	Large material size		

Table S1 Comparison between chemical transformation method and CVD/CVT method.

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