SUPPLEMENTARY MATERIAL

A. Lattice parameters of the patterned PtTe₂ monolayer with a Te vacancy

The pristine PtTe₂ system crystallizes in the CdI₂-type trigonal (1T) structure (SG P \(P \overline{3}m_1\)) is a layered material stacking along the \(z\) axis. Recently, the monolayer structure with kagome lattice formed by one Te vacancy in the \(2 \times 2\) supercell has been successfully grown \(^{18}\). The patterned PtTe₂ monolayer contains two Te layers, with 4 Te atoms in the bottom layer while 3 Te atoms in the top layer, as shown in Fig. 2(a). The corresponding lattice parameters are \(a = b = 8.1846\text{Å}, \alpha = \beta = 120^\circ\), and the thickness of the vacuum layer along \(z\) axis was set to \(30 - d_0\) Å, with \(d_0 = 2.7253\text{Å}\) denoting the distance between the bottom Te layer and the top Te layer.

| Atoms             | Wyckoff positions | Fractional coordinates          |
|-------------------|-------------------|---------------------------------|
| Pt1               | 1c                | (0.66667,0.33333,0.0)           |
| Pt2               | 3d                | (0.16667,0.33333,0.0)           |
| Te1(bottom)       | 1a                | (0.00000,0.00000,-0.04542)      |
| Te2(bottom)       | 3d                | (0.50000,0.50000,-0.04542)      |
| Te3(top)          | 3d                | (0.83333,0.66667,0.04542)       |

B. The calculated weak topological invariant \(Z_2\)

To characterize the topological properties in the patterned PtTe₂ monolayer, the weak topological invariants are calculated by the 1D Wilson loop method. Taking Te: \(5s^25p^4\) and Pt: \(5d^96s^1\) orbitals into consideration, there are \(N_e = 82\) valence electrons, resulting in \(N_e\) valence bands. From Figs. S1(a-d), the calculated weak topological invariants for \(78, 80, 82\) and \(84\) (corresponds to \(N_e - 4, N_e - 2, N_e\) and \(N_e + 2\)) occupied bands are \(Z_2 = 1, Z_2 = 1, Z_2 = 1\) and \(Z_2 = 0\), respectively. Thus, the patterned PtTe₂ monolayer with \(N_e\) valence bands is a 2D TI with \(Z_2 = 1\).

FIG. S1. (Color online) The calculated weak topological invariant \(Z_2\) for (a) 80 (b) 82 (c) 84 and (86) occupied bands, respectively.

C. Topological surface states of the patterned PtTe₂ monolayer

Exotic topological surface states serve as significant fingerprints to identify various topological phases. Based on the tight-binding (TB) model constructed with the maximally localised Wannier functions and surface Green function methods\(^{59-61}\), we have calculated the corresponding surface states to identify the nature of 2D TI.
As shown in Fig. S2(a,b), we have chosen Pt-d and Te-p orbitals as the projected bases of the wannier-based TB model, which can reproduce the DFT band structures perfectly. As expected, the helical edge states corresponds to the 2D TI can be found in the (01) edge states, as shown in Fig. S3(b) and Fig. S3(d).

FIG. S3. (Color online) The projected edge states along (01) direction (a) without SOC and (b) with SOC in the upper edge. The projected edge states along (01) direction (c) without SOC and (d) with SOC in the lower edge.
D. Releasing hydrogen in hydrogen evolution reaction

There are two different mechanisms of the releasing hydrogen in HER. One is achieved by combining a solvated proton with an adsorbed H atom $H^+ + e^- + H^* \rightarrow H_2$, known as Heyrovsky reaction, while the other one is achieved by combining two adsorbed H atoms $2H^* \rightarrow H_2$, i.e., the Tafel reaction. As shown in Fig. S4, the energy barrier in the Tafel reaction is assessed with a smaller value (0.87 eV) by the climbing-image nudged elastic band (CI-NEB) approach\cite{62}, which is 0.57 eV lower than that in the Heyrovsky reaction.

![Energy pathways of the (a) Heyrovsky and (b) Tafel reactions on PtTe1.75 monolayer to release hydrogen. The insets are the side view of the initial, transition and final states.](image)

FIG. S4. (Color online) Energy pathways of the (a) Heyrovsky and (b) Tafel reactions on PtTe$_{1.75}$ monolayer to release hydrogen. The insets are the side view of the initial, transition and final states.

E. Screening adsorption sites and the correction of Gibbs free energy

In order to screen multiple thermodynamically stable adsorption sites, we uniformly generated 100 hydrogen adsorption structures with the $\{x,y\}$ coordinates of the H atoms dispersing uniformly in the quarter of the $2 \times 2$ PtTe$_{1.75}$ supercell. After optimizing the $z$ coordinate of the adsorbed H atoms, we choose five most thermodynamically stable structures. After the fully structural optimization, we find that four of them host the same ultimate configuration (denoted as PtTe$_{1.75}$-I) with the lowest energy. While the other ultimate configuration (denoted as PtTe$_{1.75}$-II) is metastable with a 6.14 meV/atom higher energy than the PtTe$_{1.75}$-I phase.

The change of Gibbs free energy induced by hydrogen adsorption ($\Delta G_{H^*}$) can be defined as\cite{55}

$$\Delta G_{H^*} = G(H^*) - G(*) - \frac{1}{2} G(H_2)$$

$$G = E + ZPE + \int C_p dT - TS$$

(3)

where $E$ is internal Energy, ZPE is zero-point energy, $\int C_p dT$ is the correction of enthalpy, $T$ is temperature, while $S$ denotes entropy. For the adsorption structure ($H^*$), we only pay attention to the vibration contribution of the adsorbed H atom, while the correction energy of the whole PtTe$_{1.75}$ slab can be considered as unchanged before and after the absorbed process. In order to take the effect of structural size on $\Delta G_{H^*}$ into consideration, we have chosen the most thermodynamically stable PtTe$_{1.75}$ adsorption structures with $1 \times 1$, $2 \times 2$, $3 \times 3$ and $4 \times 4$ supercell. As shown in Table S2, we can find that the effect of the size is almost negligible.

F. Band structures vs. doping

Since the electronic properties and SHC of the patterned PtTe$_2$ monolayer are sensitive to the $E_F$, we can adopt different dopings at the Te-vacancy position to tune $E_F$ effectively.
TABLE S2. The Gibbs free energy correction terms of the most thermodynamically stable PtTe$_{1.75}$ adsorption structures with $1 \times 1$, $2 \times 2$, $3 \times 3$ and $4 \times 4$ supercell, including adsorption energies of hydrogen ($\Delta E_{H^*}$), the change of zero-point energy ($\Delta ZPE$), enthalpy correction ($\Delta \int C_p \, dT$), entropy correction ($\Delta TS$), and Gibbs free energy ($\Delta G_{H^*}$). All units are eV.

|       | $\Delta E_{H^*}$ | $\Delta ZPE$ | $\Delta \int C_p \, dT$ | $\Delta TS$ | $\Delta G_{H^*}$ |
|-------|------------------|--------------|--------------------------|-------------|-----------------|
| 1×1   | -0.1464          | 0.0732       | -0.0371                  | -0.1911     | 0.0809          |
| 2×2   | -0.1438          | 0.0725       | -0.0369                  | -0.1909     | 0.0826          |
| 3×3   | -0.1401          | 0.0734       | -0.0371                  | -0.1911     | 0.0874          |
| 4×4   | -0.1471          | 0.0734       | -0.0370                  | -0.1911     | 0.0803          |

As shown in Figs. S5(a-c), we can find that both the introduced Pb doping and Tl doping behave as electron dopings, which increase the $E_F$ significantly. The decreased magnitude of the $E_F$ are estimated to be 0.482232 eV and 0.256149 eV, respectively.

FIG. S5. (Color online) Band structures of the patterned PtTe$_2$ monolayer with (a) no doping (Te vacancy), (b) Tl doping, and (c) Pb doping at the Te-vacancy position.