

## Supplementary Materials

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### S1. Theoretical Methods

Modelling of the atomic structure and energetics of gas adsorption on Pt<sub>3</sub>Te<sub>4</sub> was carried out using the QUANTUM-ESPRESSO code [1] and the GGA-PBE functional with van der Waals (vdW) corrections, feasible for the studying of the adsorption of molecules on surfaces [2,3]. Energy cutoffs of 25 and 400 Ry for the plane-wave expansion of the wave functions and the charge density, respectively, and the 4 × 4 × 3 Monkhorst-Pack k-point grid for the Brillouin sampling were used [4]. For the modeling of the surface, we used slab of two Pt<sub>3</sub>Te<sub>4</sub> layers each of these layers contain PtTe<sub>2</sub> and Pt<sub>2</sub>Te<sub>2</sub> layers (see Figure 1). Note that in the slab of any number of Pt<sub>3</sub>Te<sub>4</sub> layers on one surface will be Pt<sub>2</sub>Te<sub>2</sub> layer and on opposite side PtTe<sub>2</sub>. Moreover, we also considered the presence of the Te vacancies in top layer. To imitate contribution from rigid subsurface area of bulk crystals in the modeling of the surfaces, we performed optimization of the only atomic positions. In order to take into account the contribution from flexibility of nanosheets, we performed optimization of both atomic positions and lattice parameters. The enthalpies of physical adsorption were calculated by the standard formula:

$$\Delta H_{\text{phys}} = [E_{\text{host+mol}} - (E_{\text{host}} + E_{\text{mol}})]$$

Here,  $E_{\text{host}}$  is the total energy of the surface before adsorption, and  $E_{\text{mol}}$  is the energy of the single molecules of considered species in empty box. In the case of water adsorption, we only considered adsorption from the gaseous phase. Energy of chemical adsorption is defined as difference between the total energy of the system after and before decomposition of physically adsorbed molecule. For the case of physical adsorption, we also evaluated differential Gibbs free energy the formula

$$\Delta G = \Delta H - T\Delta S$$

where  $T$  is the temperature and  $\Delta S$  is the change of entropy after formation molecule–substrate noncovalent bond, which was estimated similar to the gas  $\rightarrow$  liquid transition and hence can be evaluated by the standard formula

$$\Delta S = \Delta H_{\text{vap}}/T$$

where  $\Delta H_{\text{vap}}$  is the empirical enthalpy of vaporization.

All formulas and technical details for the calculations of the HER and OER are the same as used for the modeling of these reactions over a Pt(111) substrate [5,6].

## References

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