

## Tunable Schottky and Ohmic contacts in graphene and tellurene van der Waals heterostructures

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### I. CONFIGURATIONS OF G/TE VDWHS

The G/Te vdWHs may form multiple types of configurations depending on the approach of tellurene towards graphene. To determine the most stable structures, we consider three representative stacking configurations of graphene on tellurene in our calculations. As shown in Fig. S1, the three configurations correspond to that the topmost tellurium atoms (or most of the topmost tellurium atoms in  $\beta$ -Te) are situated directly below either: (1) carbon atoms, (2) the centers of C-C bonds, or (3) the hollow sites at the center of the C hexagonal ring, which are classified as the top (T), the bridge (B), and the hollow (H) configurations, respectively. For all three configurations, graphene and tellurene keep the optimized equilibrium interlayer distances of  $D \approx 3.594$  Å for the G/ $\alpha$ -Te vdWH and  $D \approx 3.45$  Å for the G/ $\beta$ -Te vdWH after structural relaxation. Table. S1 shows the calculated total energies per supercell of G/Te vdWHs in the T, B, and H configurations. Obviously, for both G/Te vdWHs, the H configurations have the lowest total energies and are energetically the most stable configurations.

We also examine the electronic structures of Te/G vdWHs in the T, B, and H configurations. Fig. S2 shows the projected band structures of G/Te in the three configurations. For both G/Te vdWHs, we find that all three configurations exhibit very similar band structure features.

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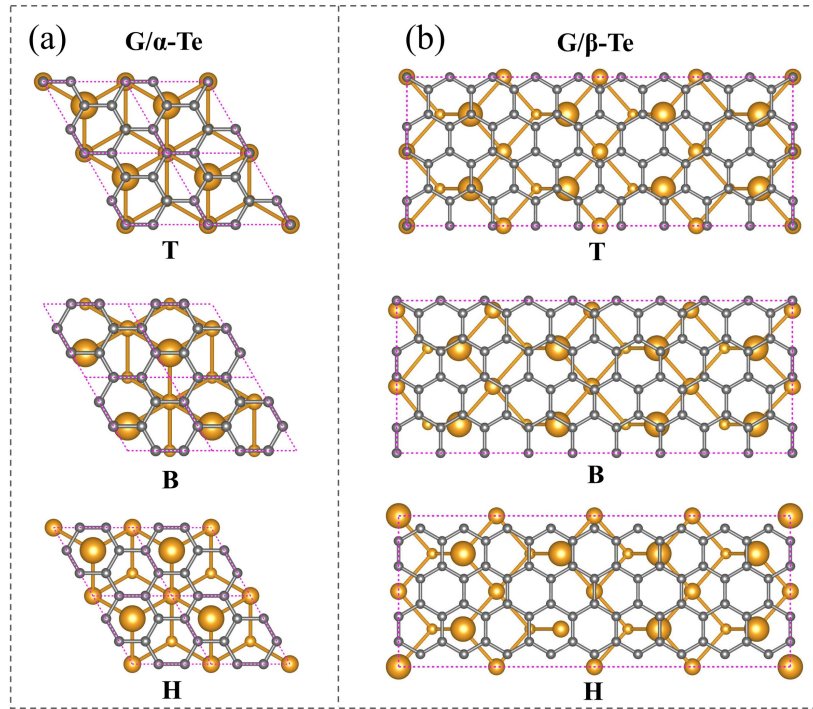


Fig. S1: Top views of the optimized structures of the top (T), bridge (B), and hollow (H) configurations for the (a)  $G/\alpha$ -Te and (b)  $G/\beta$ -Te vdWHs. These configurations are classified according to the relative position of the topmost Te atoms and graphene. The orange balls of different sizes represent the Te atoms of different positions in the vertical direction, and the largest balls represent the topmost Te atoms.

Table S1: Calculated equilibrium interlayer distance  $D$ , total energy per supercell  $E_{\text{tot}}$  of  $G/\text{Te}$  vdWHs in the T, B, and H configurations.

vdWHs	$G/\alpha$ -Te		$G/\beta$ -Te	
	$D$ ( $\text{\AA}$ )	$E_{\text{tot}}$	$D$ ( $\text{\AA}$ )	$E_{\text{tot}}$
T	3.594	-171.0471	3.456	-1608.3575
B	3.594	-171.0496	3.454	-1608.3717
H	3.594	-171.0525	3.450	-1608.3791

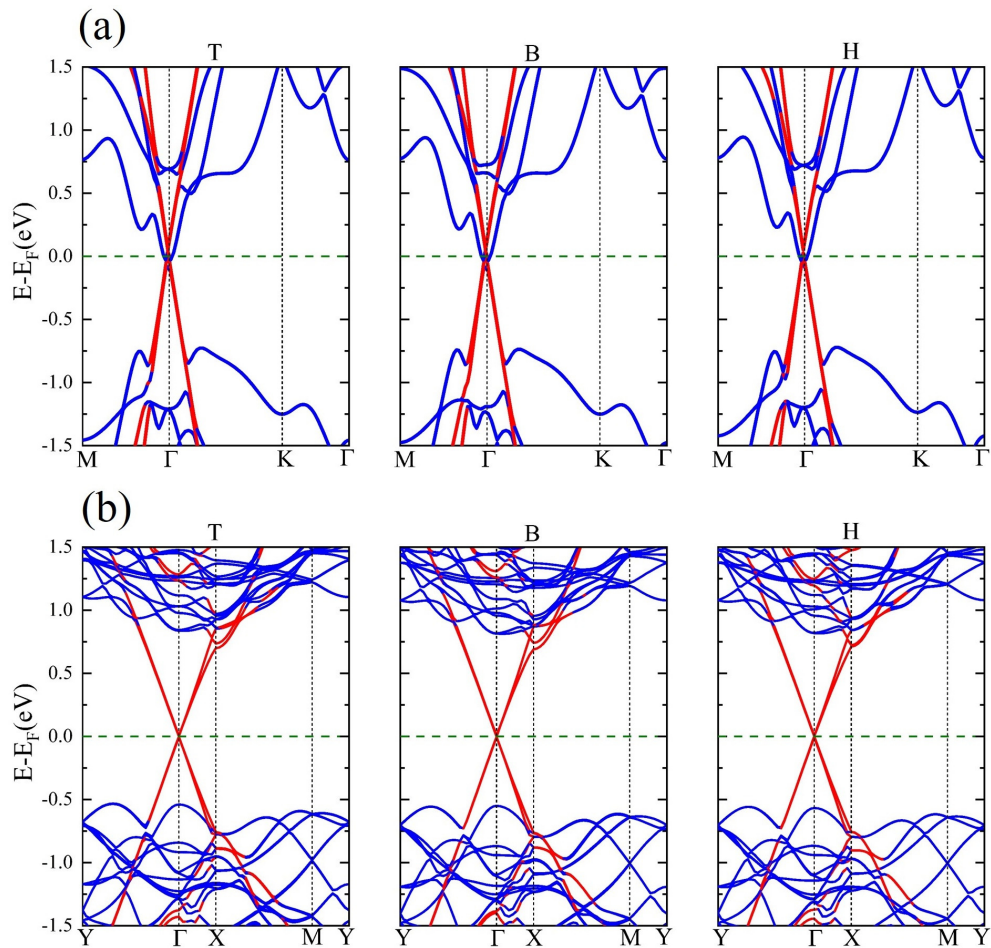


Fig. S2: Projected band structures of (a) G/ $\alpha$ -Te and (b) G/ $\beta$ -Te vdWHs in the T, B, and H configurations. The main contributions of bands from graphene and tellurene are represented by the red and blue solid lines. The Fermi levels are set to zero and marked by olive dashed lines.

## II. STABILITY OF G/TE VDWHS

To check the thermal stability of G/Te vdWHs at room temperature, we perform Ab Initio Molecular Dynamics (AIMD) simulations with different temperatures of 300 and 500 K, as shown in Fig. S3 and Fig. S4. During the AIMD simulations, the total energies of G/Te vdWHs are fluctuating in a narrow range, and no obvious structural reconstructions and separation of graphene and tellurene occur at temperatures of 300 and 500 K. Our AIMD results thus reveal that the G/Te vdWHs are thermally stable up to at least 500 K.

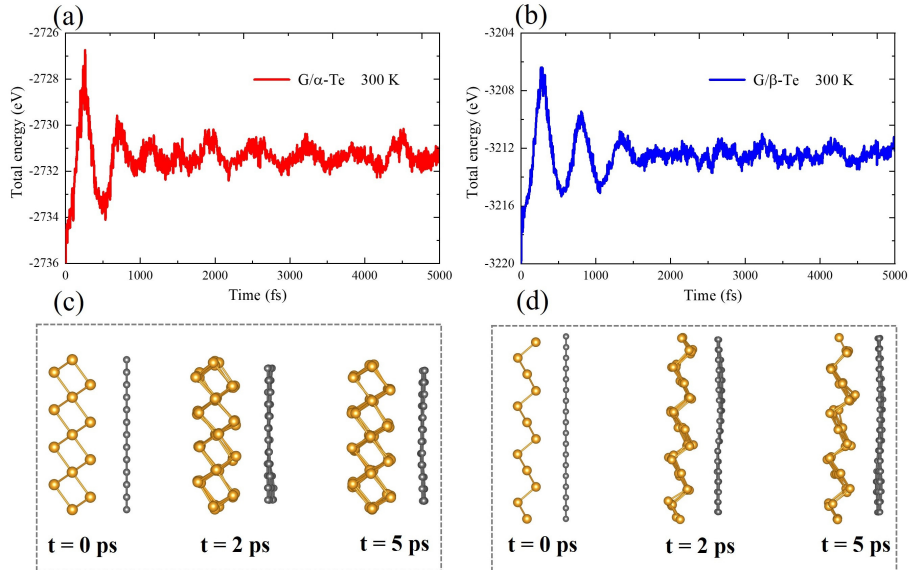


Fig. S3: Variations of the total energies of (a) G/ $\alpha$ -Te and (b) G/ $\beta$ -Te vdWHs during the AIMD simulations at the temperature of 300 K. Side views of three snapshots (t = 0, 2 and 5 ps) of (c) G/ $\alpha$ -Te and (d) G/ $\beta$ -Te vdWHs at the temperature of 300 K.

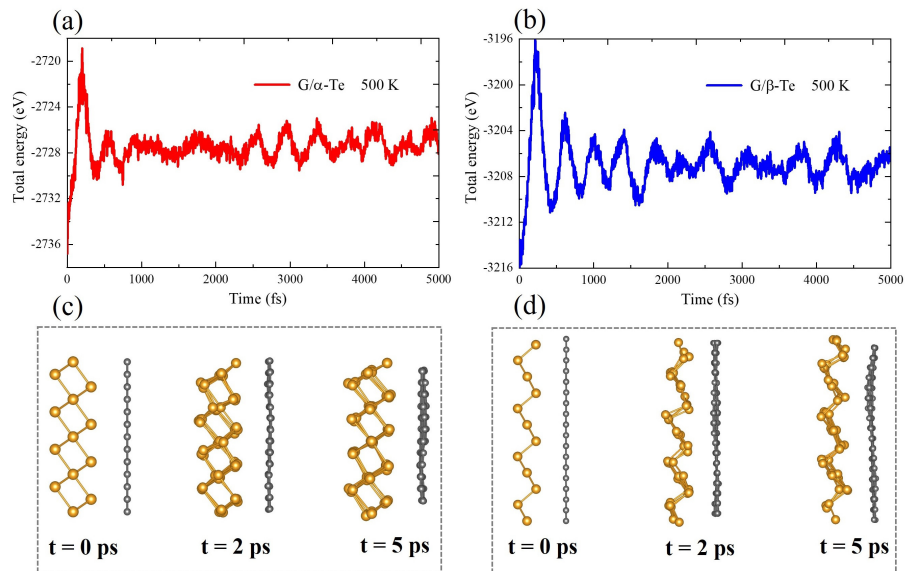


Fig. S4: Variations of the total energies of (a) G/ $\alpha$ -Te and (b) G/ $\beta$ -Te vdWHs during the AIMD simulations at the temperature of 500 K. Side views of three snapshots (t = 0, 2 and 5 ps) of (c) G/ $\alpha$ -Te and (d) G/ $\beta$ -Te vdWHs at the temperature of 500 K.

### III. TUNABILITY OF SBHS AT THE G/TE INTERFACES

To ensure the reliability of our results, we also carry out HSE06 calculations for G/Te vdWHs under external electric fields and different interlayer distances. As shown in Fig. S5 and Fig. S6, the HSE06 calculations show overall consistent tunability of Schottky barrier heights (SBHs) with that obtained by SCAN-rVV10. The main difference is that the transition of contact type displays at different values of external electric field or interlayer distance.

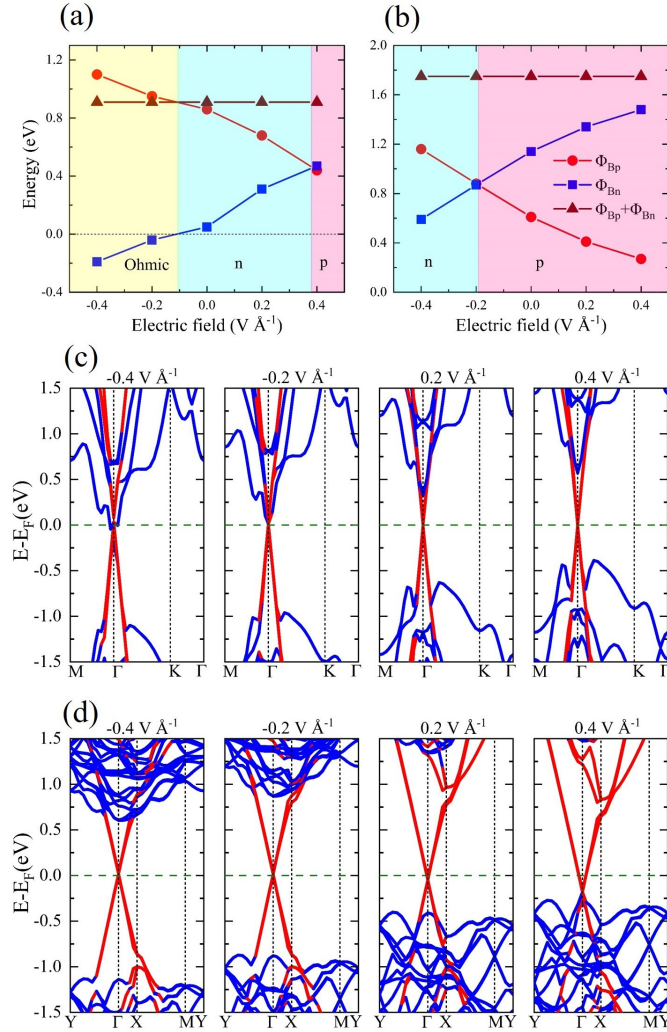


Fig. S5: HSE06 calculated Schottky barriers  $\Phi_{Bp}$ ,  $\Phi_{Bn}$ , and  $\Phi_{Bp} + \Phi_{Bn}$  in (a) G/ $\alpha$ -Te and (b) G/ $\beta$ -Te vdWHs as a function of external electric field. Projected HSE06 band structures under different electric fields ( $E_{\text{ext}} = -0.4, -0.2, 0.2, \text{ and } 0.4 \text{ V \AA}^{-1}$ ) for (c) G/ $\alpha$ -Te and (d) G/ $\beta$ -Te vdWHs. The red and blue solid lines represent the main contributions of bands from graphene and tellurene. The gray, pink, and yellow backgrounds present n-type Schottky, p-type Schottky, and Ohmic contacts, respectively. The Fermi levels are set to zero and marked by olive dashed lines.

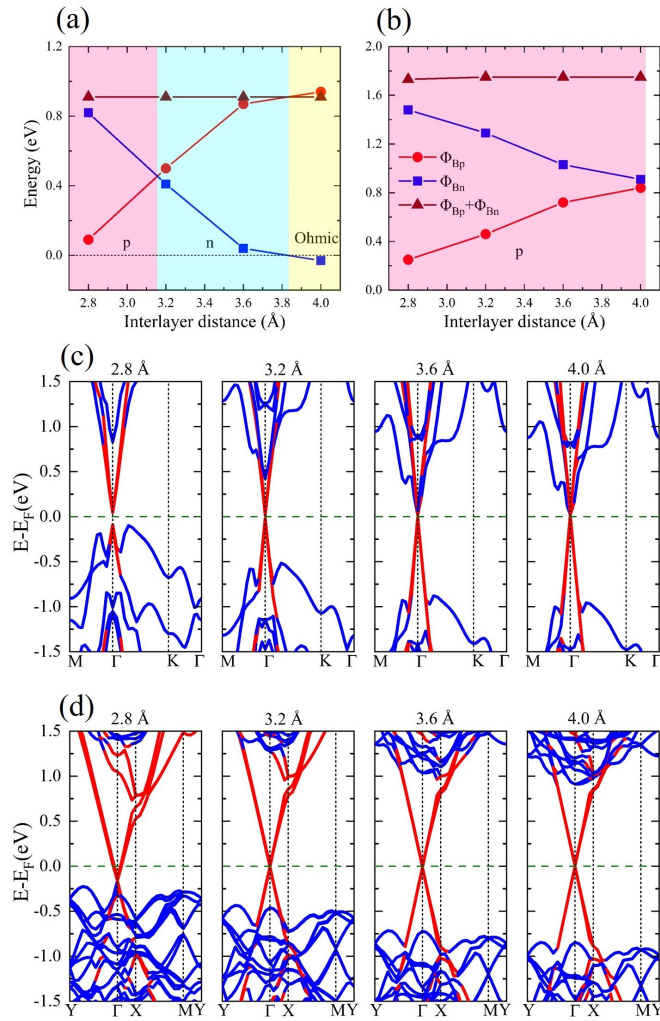


Fig. S6: HSE06 calculated Schottky barriers  $\Phi_{Bp}$ ,  $\Phi_{Bn}$ , and  $\Phi_{Bp} + \Phi_{Bn}$  in (a) G/ $\alpha$ -Te and (b) G/ $\beta$ -Te vdWHs as a function of interlayer distance. Projected HSE06 band structures at different interlayer distances ( $D = 2.8, 3.2, 3.6,$  and  $4.0 \text{ \AA}$ ) for (c) G/ $\alpha$ -Te and (d) G/ $\beta$ -Te vdWHs. The red and blue solid lines represent the main contributions of bands from graphene and tellurene. The gray, pink and yellow backgrounds present n-type Schottky, p-type Schottky, and Ohmic contacts, respectively. The Fermi levels are set to zero and marked by olive dashed lines.