Electronic Supplementary Information (ESI)

Tunable electronic properties and Schottky barrier in Graphene/WSe_2 heterostructure under out-of-plane strain and electric field

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1. The phonon dispersion curves and free energy fluctuations of WSe_2 monolayer are depicted in Fig. S1. As can be seen from Fig. S1(a), there are three acoustic branches in the phonon spectrum of WSe_2 monolayer, in which the longitudinal acoustic branch (LA) and the transverse acoustic branch (TA) vibrated in-plane have approximately a linear dispersion relation around the G point and higher frequency than the out-of-plane acoustic waves (ZA). The transverse optical branch (TO_1) and longitudinal optical modes (LO_1) near 173 cm^{-1} have weak coupling with charge carriers. TO_2 and LO_2 modes describe the case where W atoms vibrate in the opposite direction to Se atoms. In addition, the fairly small LO-TO splitting values are calculated to be 0.002 cm^{-1} and 2.484 cm^{-1} at the high symmetry G point with and without SEDC, respectively, indicating a polarization field inside WSe_2 monolayer. The out-of-plane transverse optical branches ZO_1 and ZO_2 describe that the two Se atomic layers vibrate in the same and opposite directions, respectively. The phonon dispersion of WSe_2 monolayer is similar to that of MoS_2 monolayer.[1] Fig. S1(b) shows the free energy fluctuations of WSe_2 monolayer at 273 K and 800 K. The y-coordinate values are defined by $\Delta E = E - E_0$, where E and E_0 are the energy at set temperatures and 0 K, respectively.

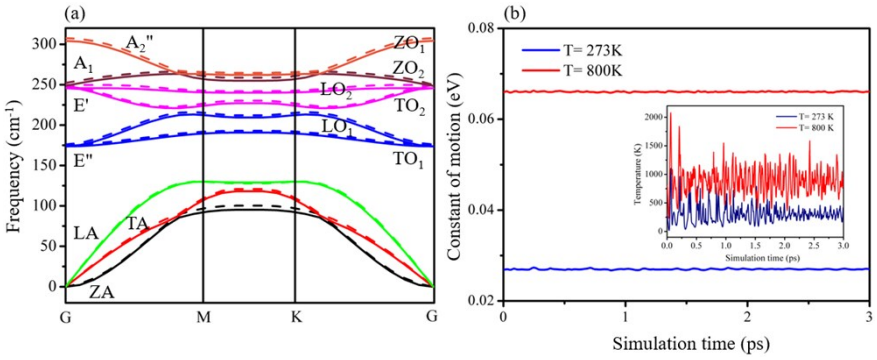

Figure S1. (a) The phonon dispersion and (b) the free energy fluctuation with different temperatures of WSe_2 monolayer. The dotted and solid lines represent the simulations with and without SEDC scheme in (a), respectively.

2. The out-of-plane strain perpendicular to the graphene/WSe_2 interface as shown in Fig. S1(a) is applied by changing the interlayer distance. For the experiment, it is an important subject to realize the precise control of interlayer distance. Heteroatom doping, metal ion intercalation, small molecule/polymer intercalation and metal atom intercalation are the common approaches to effectively modulate the interlayer distance.[2] Fig. S2(b) shows that the optimized interlayer distance is 3.59 Å at the equilibrium state. It can be seen from Fig. S2(c) that when the interlayer distance is greater than 3.4 Å, the band gap will not be affected by the interlayer distance. In Fig. S2(d), the position of π bands of graphene remain unchanged and the Dirac cone angles formed by the π and π* bands gradually decrease with increasing the interlayer distance. The band position of graphene changed. The π* band is closest to the Fermi level and the opened band gap of the graphene/WSe_2 heterostructure is 38 meV when the interlayer distance is 2.6 Å.

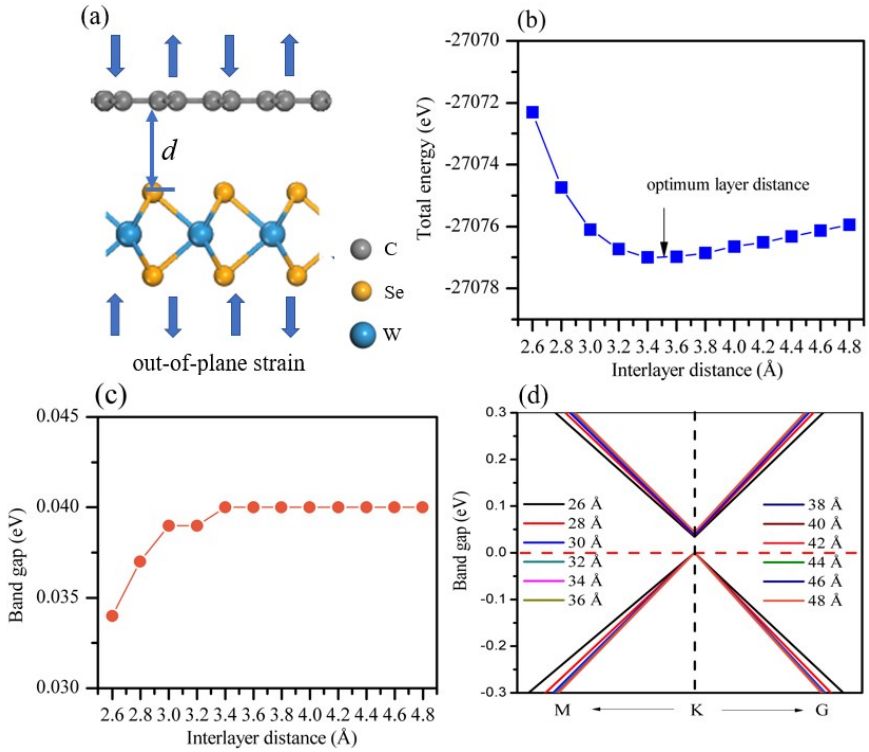

Figure S2. (a) The schematic diagram of the graphene/WSe_2 heterostructure by changing the out-of-plane strain. Calculated (b) total energy; (c) band gap and (d) band structure near Fermi level of graphene/WSe_2 heterostructure.

3. The direction of the applied electric field and the band gap of the graphene/WSe_2 heterostructure under different electric fields are shown in Fig. S3(a) and Fig. S3(b), respectively. It can be seen that when the external electric fields are -0.6~-0.4 V/Å

and +0.5~+0.6 V/Å, the band gap of the heterostructure is 0 eV, showing an Ohmic contact behavior. The Fig. S3(c) and S3(d) show the energy dispersion relation between near Dirac point of Fermi level under different external electric fields. When the electric field is between -0.6 V/Å and -0.4 V/Å, the π and π* bands of graphene are located below the Fermi level. In contrast, when the electric field is applied at +0.5 ~ +0.6 V/Å, the π and π* bands of graphene is located above the Fermi level. Therefore, the opened band gap of the graphene/WSe_2 heterostructure at the Dirac point is sensitive to its lattice symmetry and are affected by the interlayer distance and electric field.

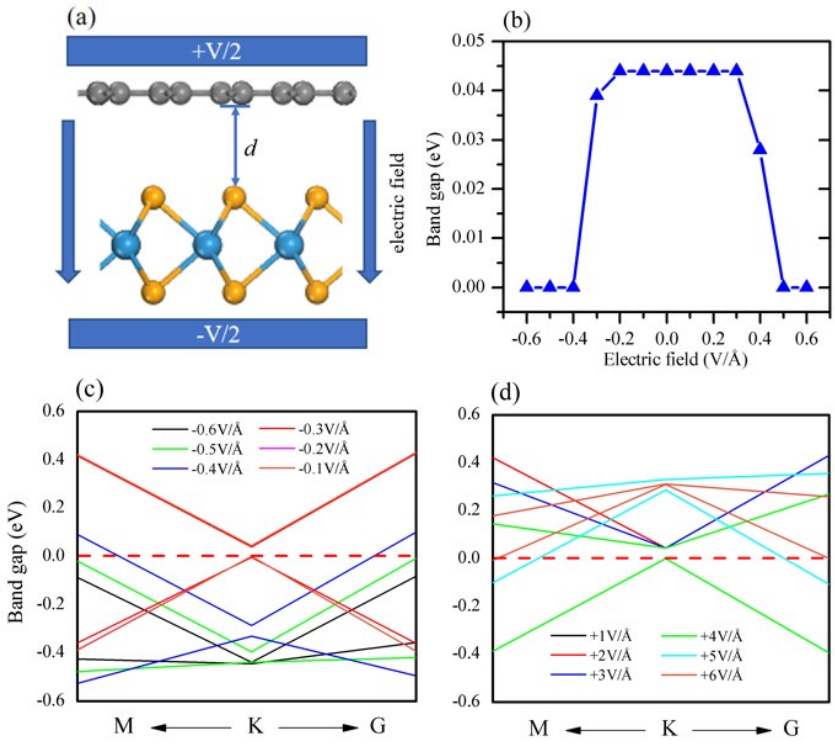

Figure S3. (a) The schematic diagram of the graphene/WSe_2 heterostructure by changing the electric field. Calculated band gap (b); band structure near Fermi level of graphene/WSe_2 heterostructure under negative electric field (c) and under positive electric field (d). The positive direction of the applied external electric field is from graphene to WSe_2 monolayer.

[1] L. F. Huang, P. L. Gong, Z. Zhi, *Physical Review B*, 2014, **90**, 045409

[2] Y. Xue, Q. Zhang, W. Wang, et al., *Advanced Energy Materials*, 2017, **7**, 1602684.