Supporting Information

Design of Phosphorene/Graphene Heterojunctions for High and Tunable Interfacial Thermal Conductance

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S1. P-G heterostructures for DFT calculations.

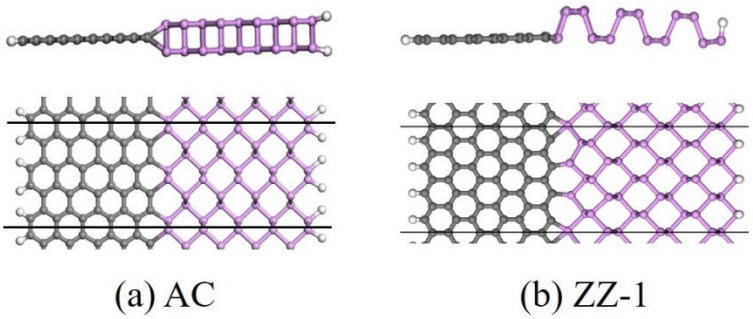

Figure S1. P-G heterostructures with (a) AC interface and (b) ZZ-1 interface used in the DFT calculations. The white, pink and gray circles denote H, P and C atoms, respectively.

S2. The Phonon dispersions of graphene and phosphorene in AC and ZZ-1 heterostructures.

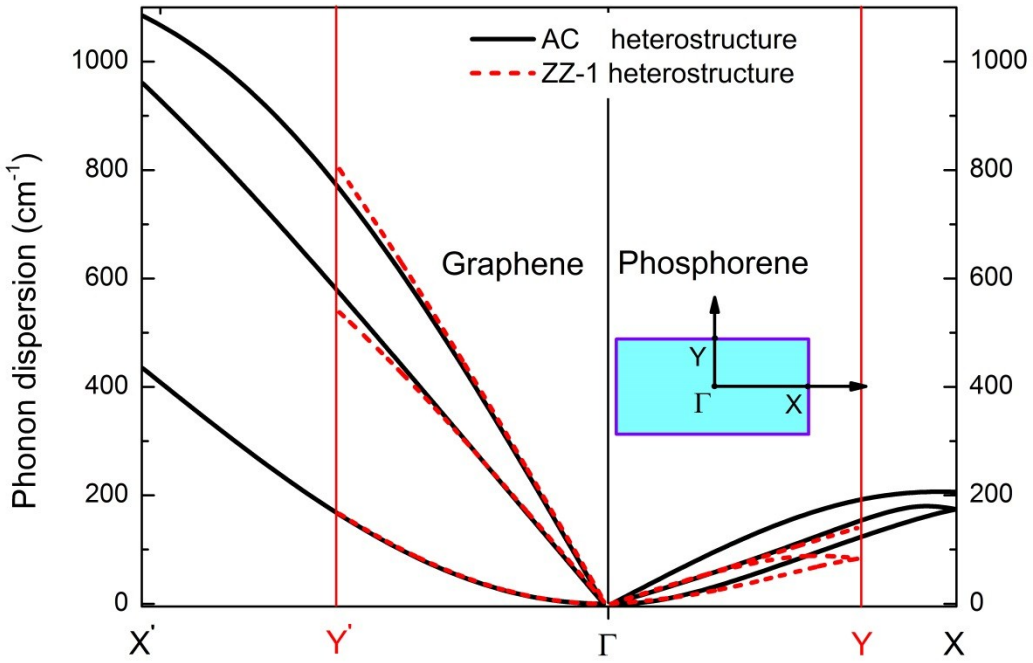

Figure S2. Phonon dispersions of graphene and phosphorene in AC and ZZ-1 heterostructures, respectively. The insert shows the Brillouin zone corresponding to the orthorhombic unit cell.

S3. The method for the calculation of stress distributions along P-G heterostructures.

Stress field along the P-G heterostructures is obtained using MD calculation.[1] The stress tensor σ_i for atom *i* is obtained by the following formula,

$$\sigma_i = \frac{1}{V_i}\left(- m_i v_i \otimes v_i + \frac{1}{2}\sum_{j \neq i} F_{ij} \otimes r_{ij} \right), \quad \text{(S1)}$$

where V_i is the volume of atom *i*, which is calculated based on the covalent radius of the atom (1.07Å for P atom, and 0.7 Å for C atom); v_i is the velocity of atom *i*; r_{ij} is the distance between atom *i* and atom *j*; F_{ij} is the interaction force between atom *i* and atom *j*. The stress tensor for atom *i* has 6 components, that is, *xx*, *yy*, *zz*, *xy*, *xz*, *yz*. They can be calculated by the formula below (Eq.S2). Here, *a* and *b* take on the values of *x*, *y*, *z* to generate the 6 components of the symmetric tensor:[1]

$$\sigma_i^{ab} = \frac{1}{V_i}\left(- m_i v_i^a v_i^b + \frac{1}{2}\sum_{j \neq i} F_{ij}^a r_{ij}^b \right). \quad \text{(S2)}$$

S4. C-P bond length distribution.

The radial distribution functions of C-P bond length at the AC and ZZ-1 interfaces are calculated under different strains, as shown in Figure S3. A single peak in Figure S3(a) indicates that only one kind of C-P bond uniformly distributed with the same length at the AC interface. Two peaks in Figure S3(b) indicate that there are two kinds of C-P bonds existing at the ZZ-1 interface. It is found that the C-P bond length at AC interface is enlarged with increasing the tensile strain; however, the bond length at ZZ-1 interface is changed only slightly.

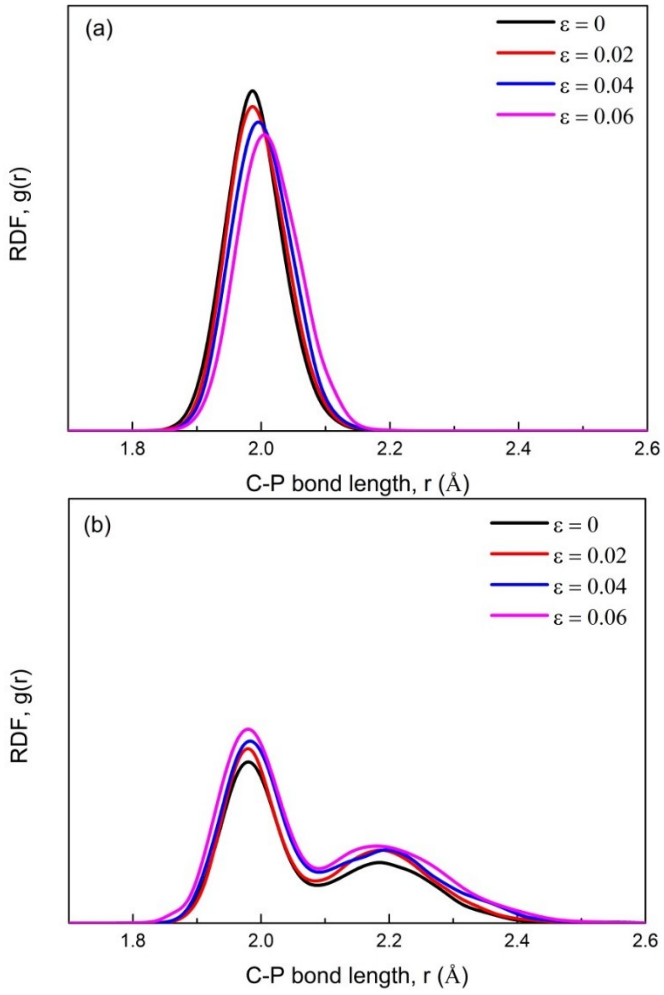

Figure S3. Radial distribution functions (RDFs) of C-P bond length at the C-P interfaces. (a) AC interface; and (b) ZZ-1 interface.

S5. The overlap of PDOS of graphene and phosphorene in AC and ZZ-1 P-G heterostructures.

Table S1. Overlap of PDOS of graphene and phosphorene at AC and ZZ-1 interfaces.

	ε = 0.0%	ε = 2.5%	ε = 4.0%
S_{AC}	0.2702	0.2716	0.2800
$S_{ZZ\text{-}1}$	0.2702	0.2643	0.2638

References

1 S. J. Plimpton *J. Comput. Phys*. 1995, **117**, 1–19.