Supporting Information:

Borophosphene: a New Anisotropic Dirac Cone Monolayer with High Fermi Velocity and Unique Feature of Self-doping

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The details of the in-house global minimum structure search code (Genetic algorithm)

The genetic algorithm (GA) is implemented to find the global minimum of the 2D BP monolayer. The implementation is shown in Fig. S1. During the iterations, each generation contains 10 structures. The initial structure is generated with random arrangement of B and P atoms constrained in a plane. In the GA used here, 8 out of 10 parent structures are chosen for crossing in order to generate child generation, with the remaining 2 structures passed to the child generation unchanged. Within the 8 structures, eight pairs are picked based on the energy of each structure: structures with lower energies (energies are from calculation of last generation) are more likely to be chosen for crossing. The probability of choosing the specific structure i is calculated as $\exp(-\lambda*(E_i-E_{min})/(E_{max}-E_{min}))$, where E_i, E_{min}, and E_{max} are the energy of structure i, lowest energy of the parent population, and highest energy of the population, respectively. $\lambda=2$ is obtained from a reference [Darby, S.; Mortimer-Jones, T. V.; Johnston, R. L.; Roberts, C. *J. Chem. Phys.* **2002**, *116*, 1536]. Once a pair of parent structures is chosen, an *x*-*y* in-plane rotation of a random angle is performed for each structure. It is followed by arranging all the atoms of a structure into a one dimensional array based on their *x*-direction coordinate, such two arrays from the two structures are crossed and exchange part of their atoms and form new child structure. During the crossing procedure, the atomic species including number of atoms of each species in the child structure need to be consistent to their parents. After the crossing, DFT relaxation is

performed to obtain the relaxed structures of the population. Mutation is also performed for each structure after the crossing with a probability around 0.1. There are different schemes of the mutation, but we find that exchange two regions of atoms (each regions contains multiple atoms) of the structure are mostly efficient to find the global minimum. The energies of the population along with the generations are shown in Figure S2 (a). Starting from the third generation, the lowest energy of the population becomes unchanged. The lowest two structures correspond to α_1 and α_2. The structures with higher energies are found to have quite similar atomic arrangement to α_2 but with distortions.

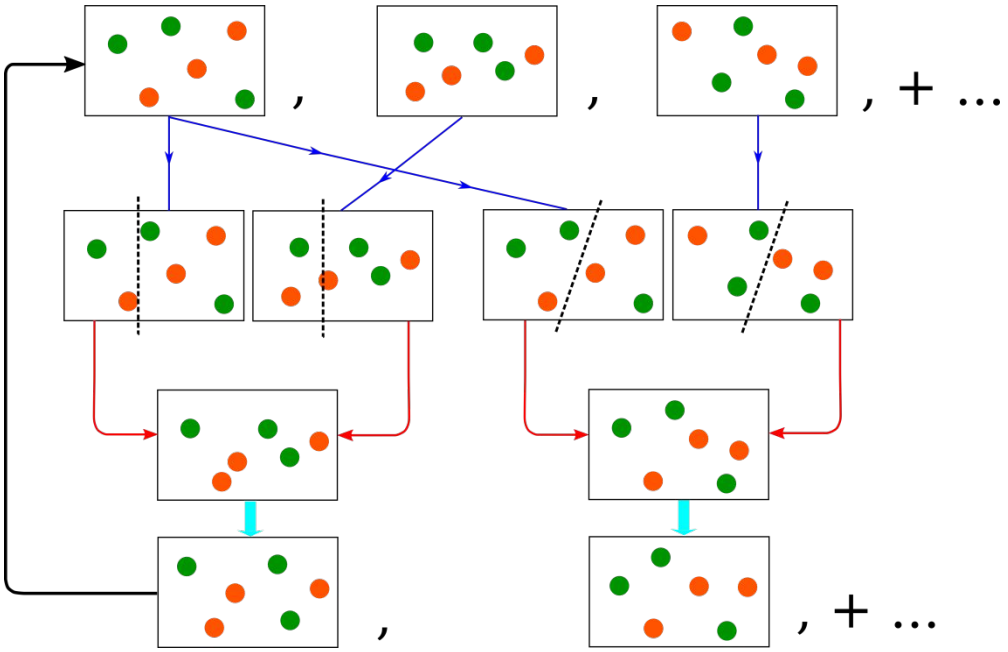

Figure S1.The process of the in-house global minimum structure search code. By picking up two structures from the parent population (blue arrow) and cutting each structure (black dotted line) described in the text, the two structures are combined into a new structure (red arrow) as the child. The child structures are further relaxed by DFT (cyan arrow) to form a child population.

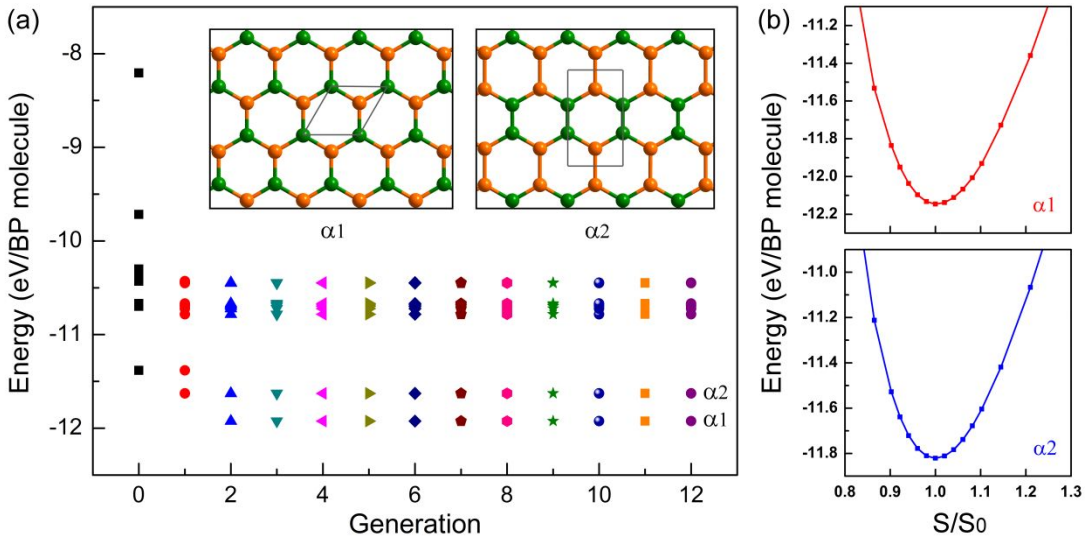

Figure S2. Energy diagrams of 2D BP monolayers against (a) the generation of Genetic Algorithms and (b) in-plane strain. Insets show the optimized geometrical structures of BP monolayers searched by using the Genetic Algorithms method. Green and orange balls denote B and P atoms.

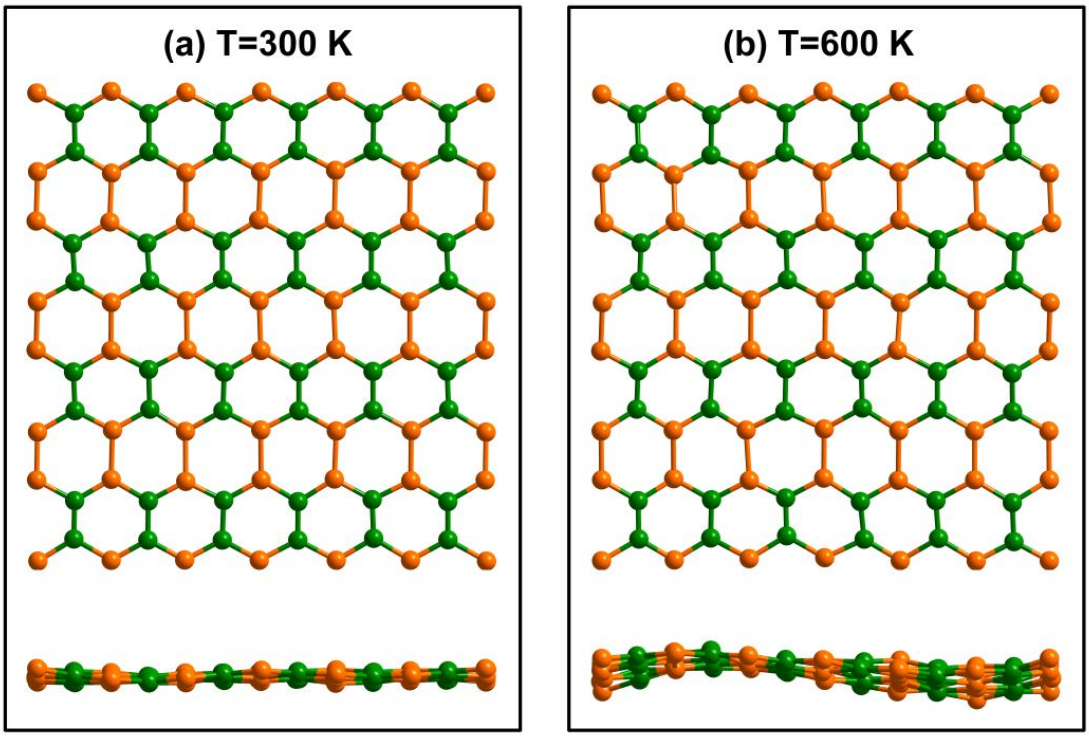

Figure S3. Snapshots of borophosphene at the temperatures of (a) 300 and (b) 600 K after 10 ps *ab initio* MD simulations.

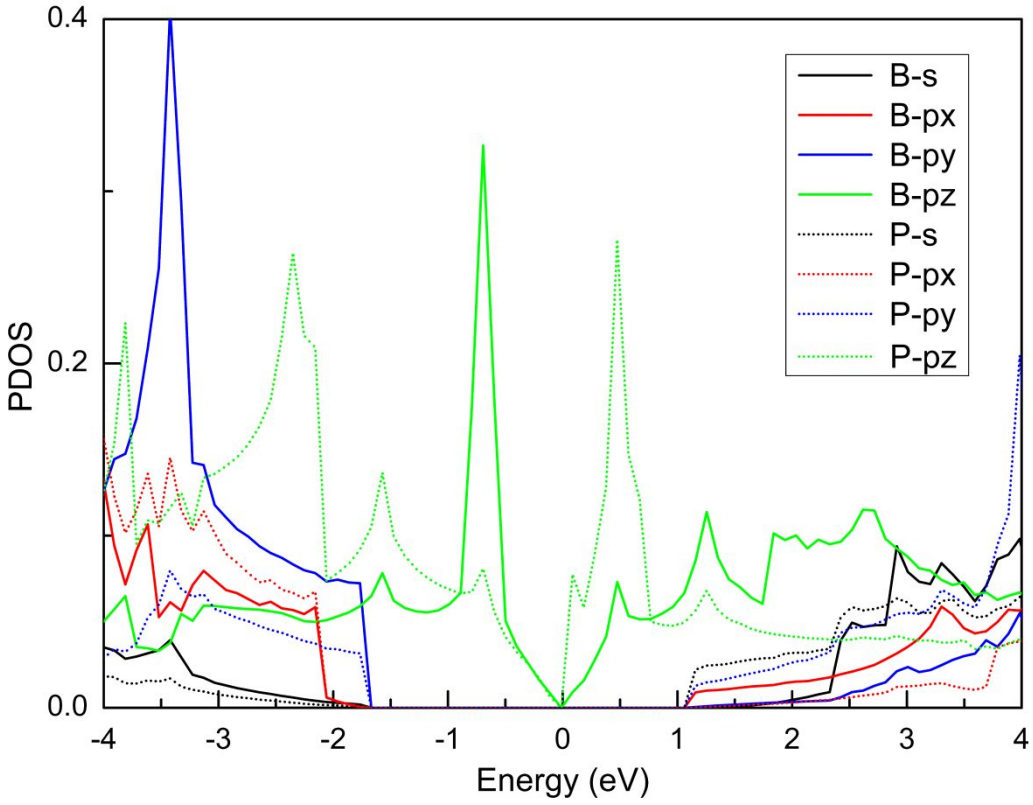

Figure S4. Partial density of state (PDOS) of borophosphene.

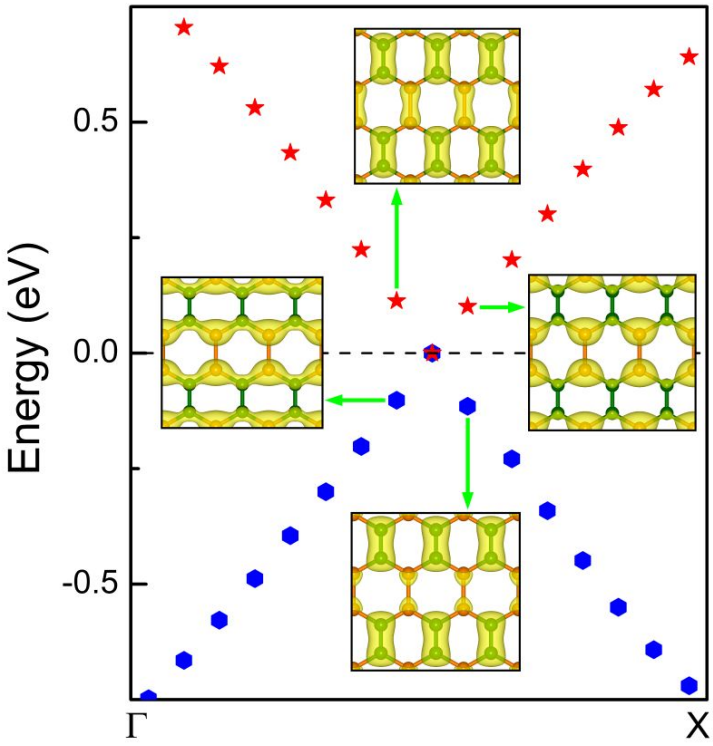

Figure S5. Spatial charge density of borophosphene around the Dirac cone.

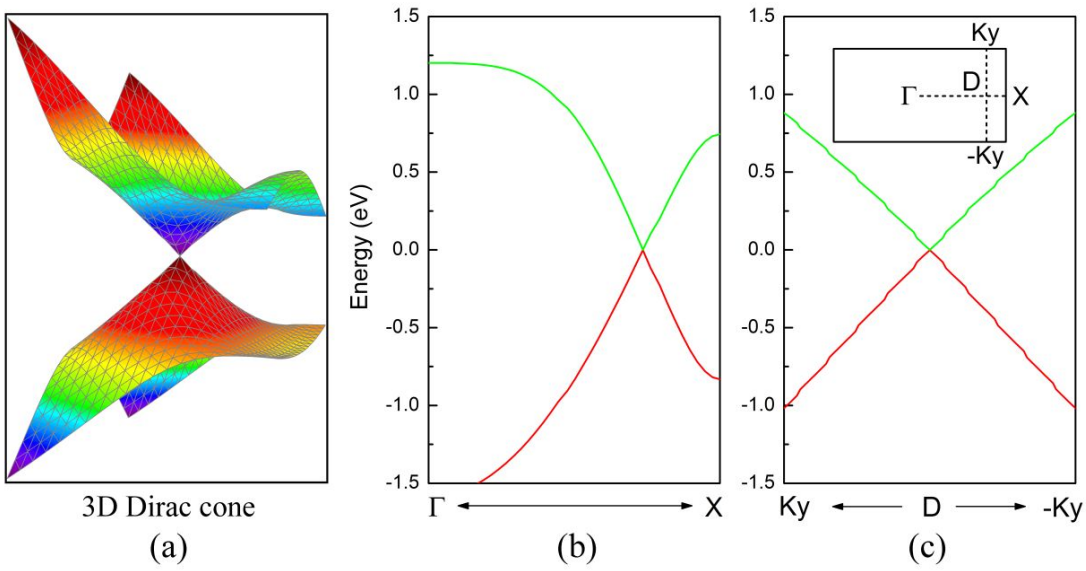

Figure S6. Calculated Dirac cones of borophosphene: (a) 3D and 2D along (b) *x* and (c) *y* axes. The inset shows the calculated *k*-point path.

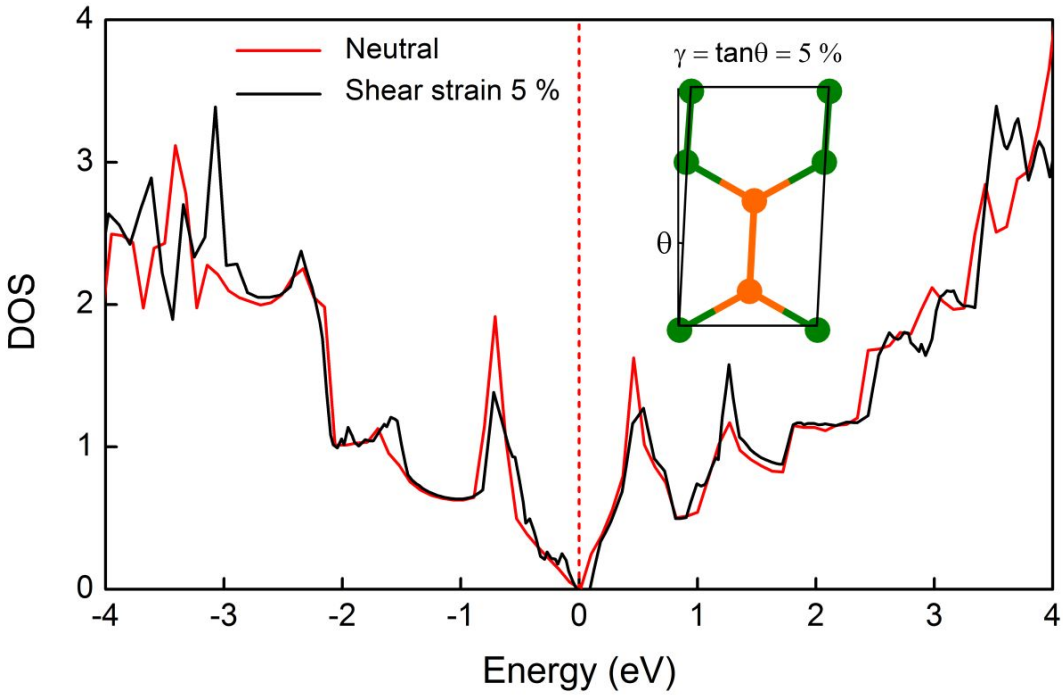

Figure S7. The calculated DOS of borophosphene under neutral and shear (5 %) strains. Inset is a schematic diagram of in-plane shear strain. It is found that the borophosphene can open a small band gap under a shear strain of 5 %, not a uniaxial or biaxial strain.

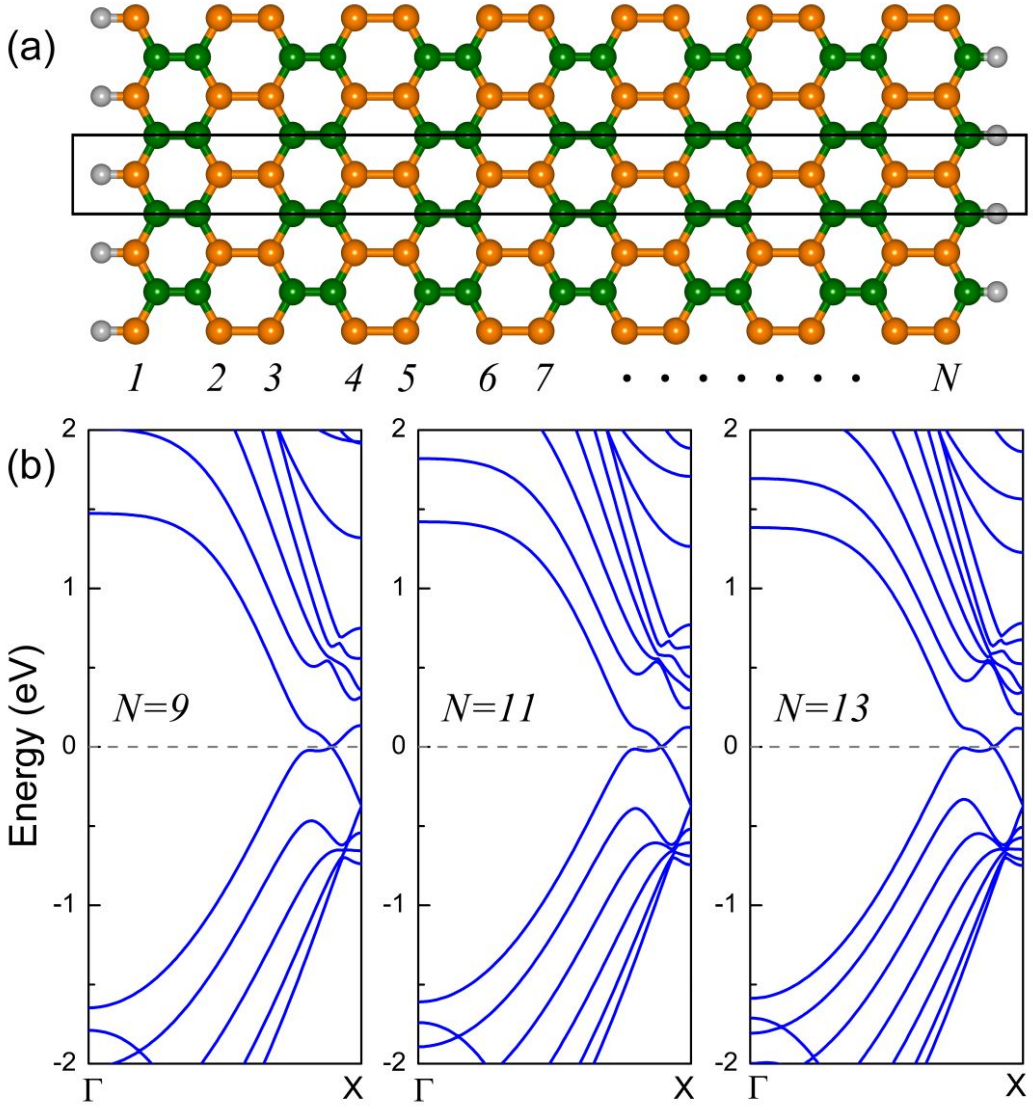

Figure S8. (a) Structural model and (b) electronic band structure of 1D zigzag nanoribbons cut from the borophosphene.

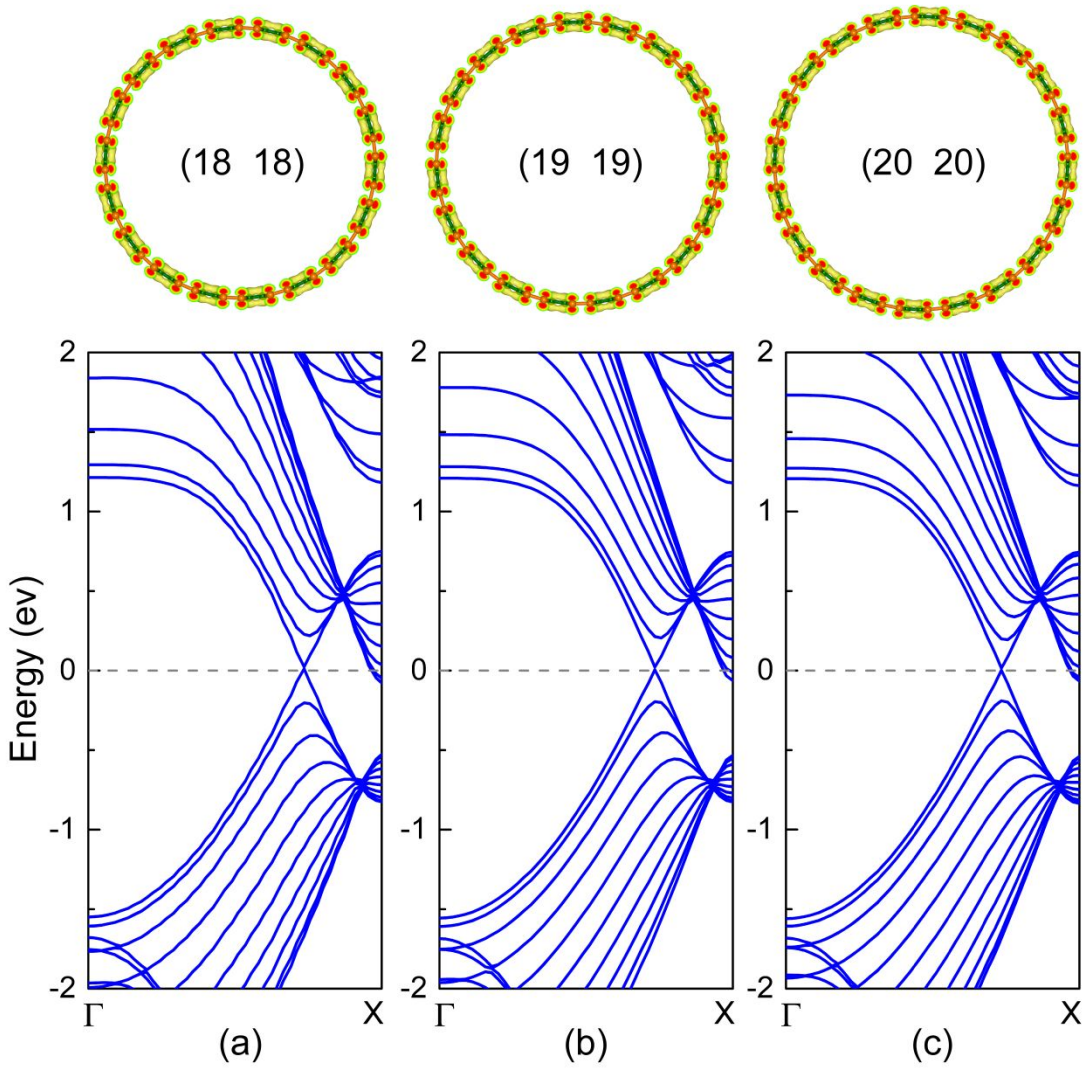

Figure S9. Electronic band structure of 1D armchair nanotubes rolled from the borophosphene: (a) (18 18), (b) (19 19), and (c) (20 20). Upper panel shows the spatial charge density around the Dirac cone.