Electronic Supplementary Material (ESI) for Nanoscale.
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Supplementary Materials for

Carbon Phosphide Monolayer with Superior Carrier Mobility

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1. Prediction of CP monolayer structures by the particle swarm optimization

The CP monolayer structures are predicted using the particle swarm optimization (PSO) method as implemented in CALYPSO code [1-4]. PSO is a method for multidimensional optimization which is inspired by the social behavior of birds flocking [2]. It has been successfully utilized to predict the crystal structures of lithium-boron compounds at high pressure [5] and other 2D materials [6]. In our case, the CP monolayers with different stoichiometric compositions are considered for calculations. The number of structures (e.g., population) that produced at each step is set to 24, and the number of CALYPSO steps (e.g., generation) is fixed to 20. The number of atoms in the unit cells is up to 10. The required structural relaxation was performed using the Perdew-Burke-Ernzerhof (PBE) [7] exchange correlation functional to density functional theory (DFT).

Figure S1 shows the cohesive energies and corresponding structures of $C_{1-x}P_x$ predicted using CALYPSO. Note the cohesive energies of all the structures are lower than graphene, but higher than phosphorene depending on their composition (Figure S1 (a)), which suggests CP monolayers with different compositions are possible to be synthesized in experiments by controlling the ratio of P/C. The details of CALYPSO steps for CP monolayers with x=0.5 are shown in Figure S2(a). The lowest energy structures (Figure S2(b)) for which are found to be comprised of 3-fold coordinated C and 2-fold coordinated P atoms, however, our phonon dispersion calculations suggest these structures with 2-fold coordinated P are not stable. The second lowest energy structures (Figure S2(c)) are comprised of 3-fold coordinated C and 3-fold coordinated P atoms (α-CP, β-CP, and γ-CP) which are found to be dynamically stable.

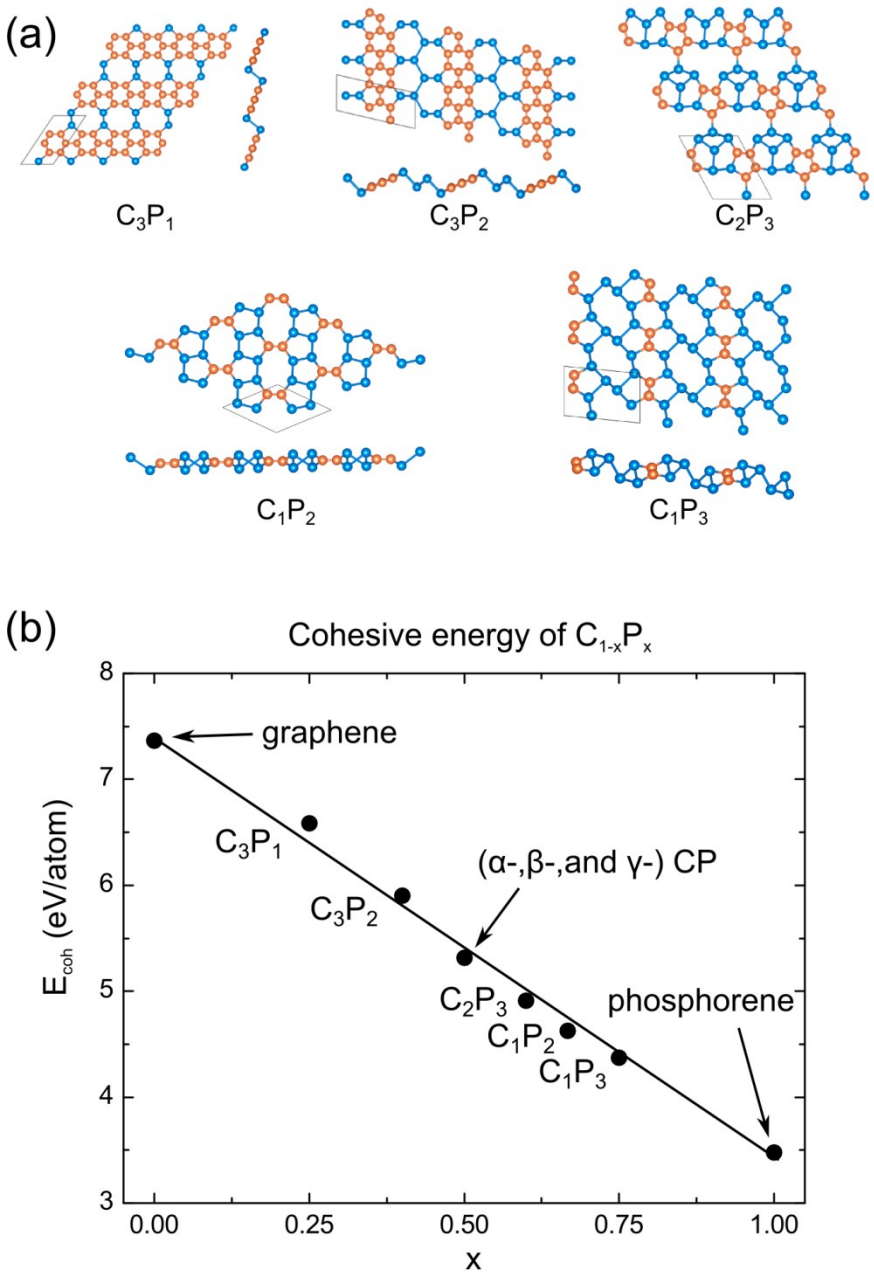

Figure S1. Calculated cohesive energy (a) and the corresponding structures (b) for CP monolayers with various stoichiometric compositions obtained using CALYPSO.

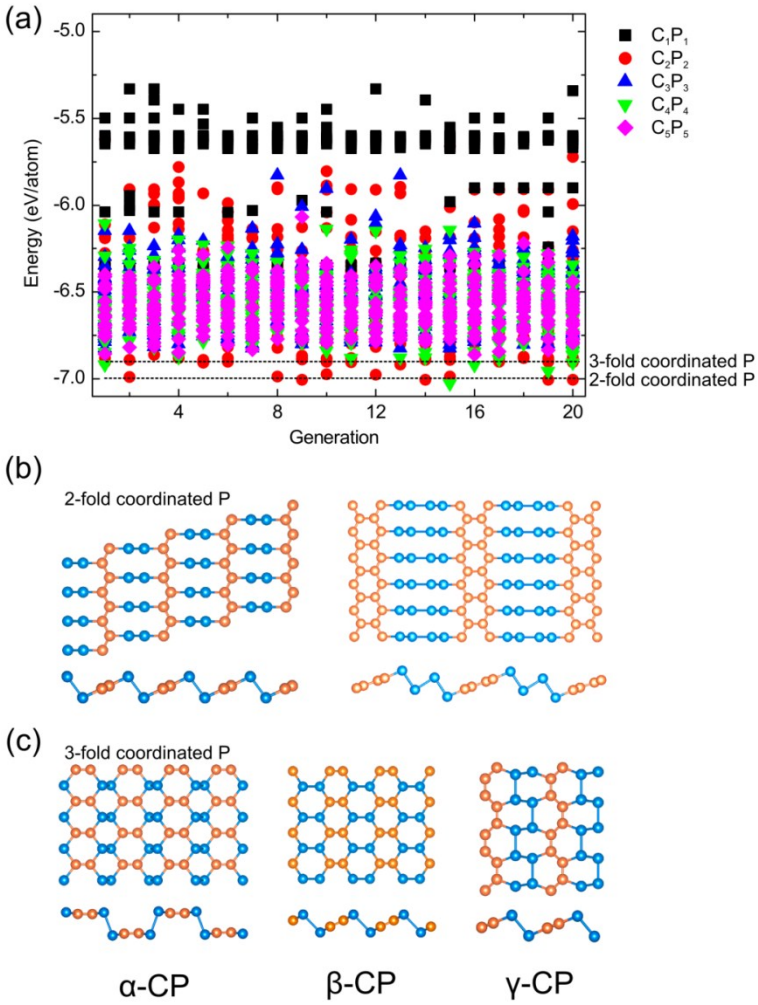

Figure S2. Stoichiometric CP monolayer (x=0.5): (a) history of CALYPSO steps, (b) structures with 2-fold coordinated P atoms, and (c) structures with 3-fold coordinated P atoms.

2. Mechanical and electronic properties of α-CP, β-CP, and γ-CP

Table S1. Calculated structural parameters of CP monolayers with the DFT-D2 functional form.

	a_1 (Å)	a_2 (Å)	$R_{C\text{-}C}$ (Å)	$R_{C\text{-}P}$ (Å)	$R_{P\text{-}P}$ (Å)	θ (°)	*Cohesive energy (eV/atom)*
α-CP	8.55	2.92	1.36	1.83	2.32	96.98	5.40
β-CP	4.70	2.91	1.37	1.82	2.33	97.59	5.41
γ-CP	4.80	5.60	1.45, 1.43	1.82	2.30, 2.17	103.08	5.43

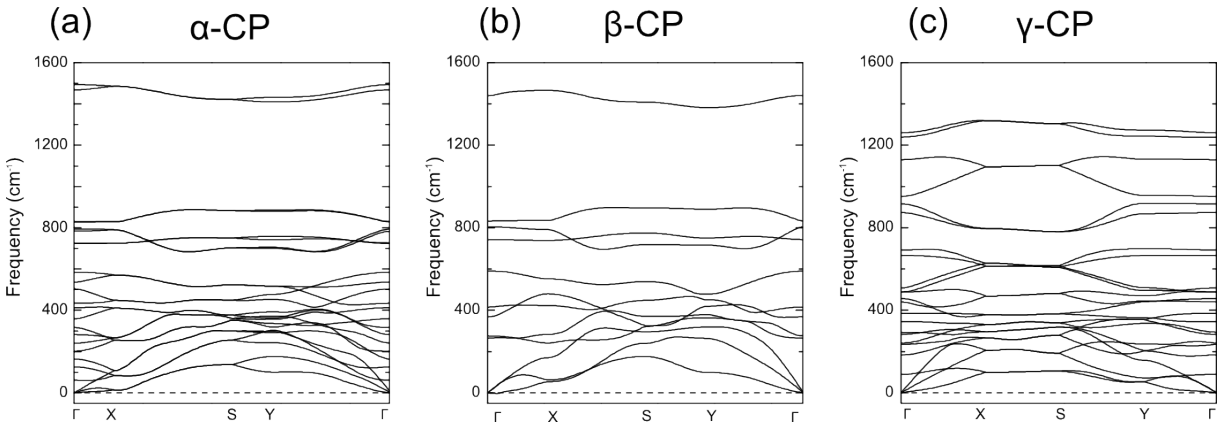

Figure S3. The phonon dispersion for (a) α-CP, (b) β-CP, and (c) γ-CP calculated with DFT-D2 functional form.

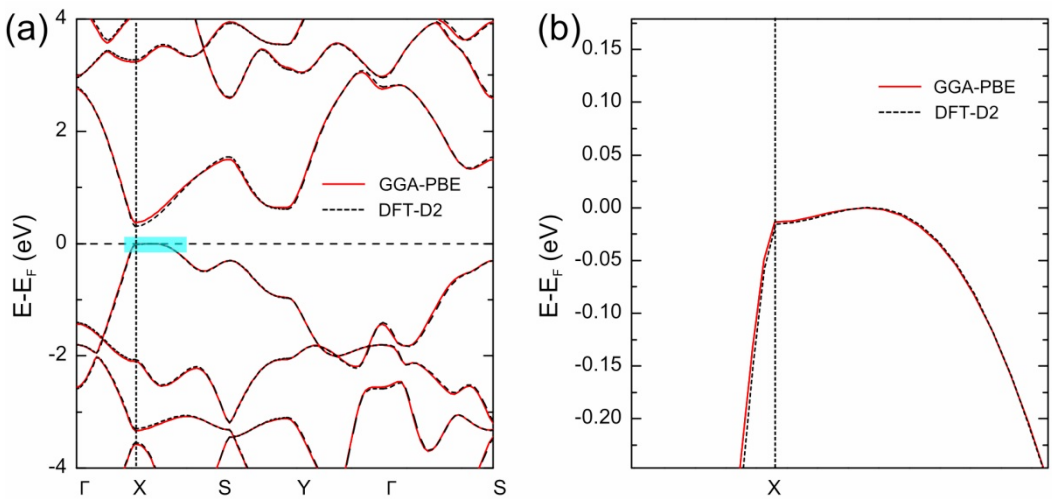

Figure S4. (a) Calculated band structure for β-CP using DFT-PBE and DFT-D2 functional form, (b) Zoomed figure for the highlighted region in (a).

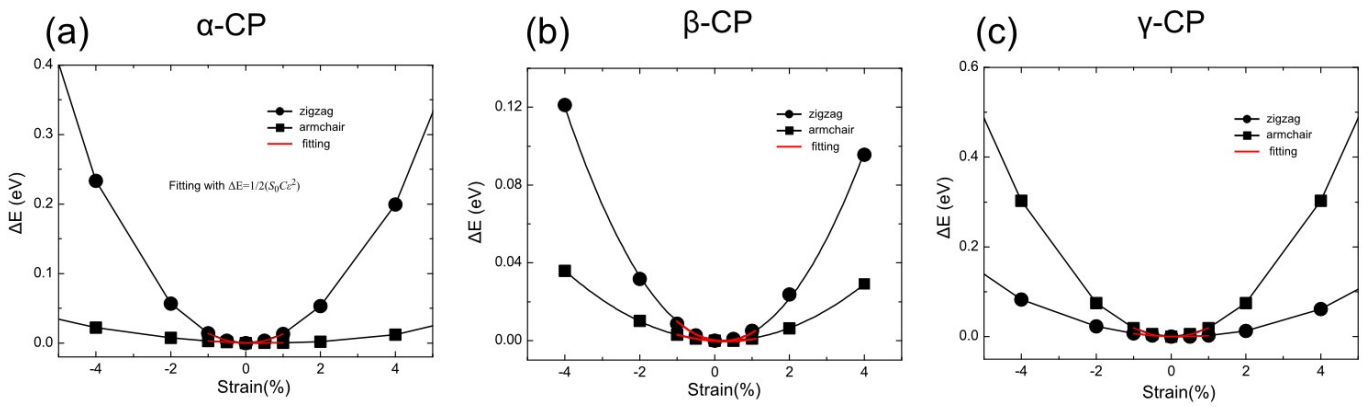

Figure S5. Strain energy curves along the armchair and zigzag directions for (a) α-CP, (b) β-CP, and (c) γ-CP. The curves are fitted within the strain range of -1% to 1% to get the in-plane stiffness constant C. S_0 the area of the unit cell at equilibrium, ε is the strain, and Δ*E* is the change of total energy compared to the equilibrium system.

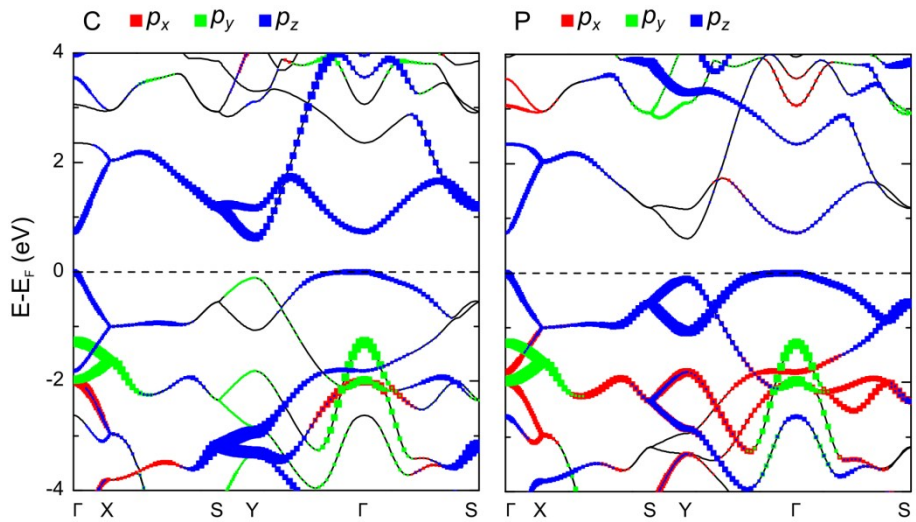

Figure S6. Atomic projected band structure associated with C atoms (left panel) and P atoms (right panel) for α-CP monolayer.

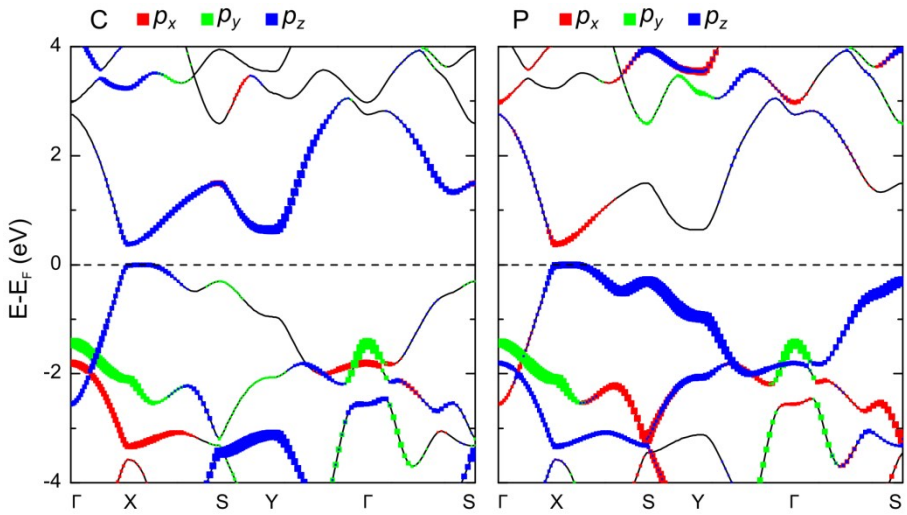

Figure S7. Atomic projected band structure associated with C atoms (left panel) and P atoms (right panel) for β-CP monolayer.

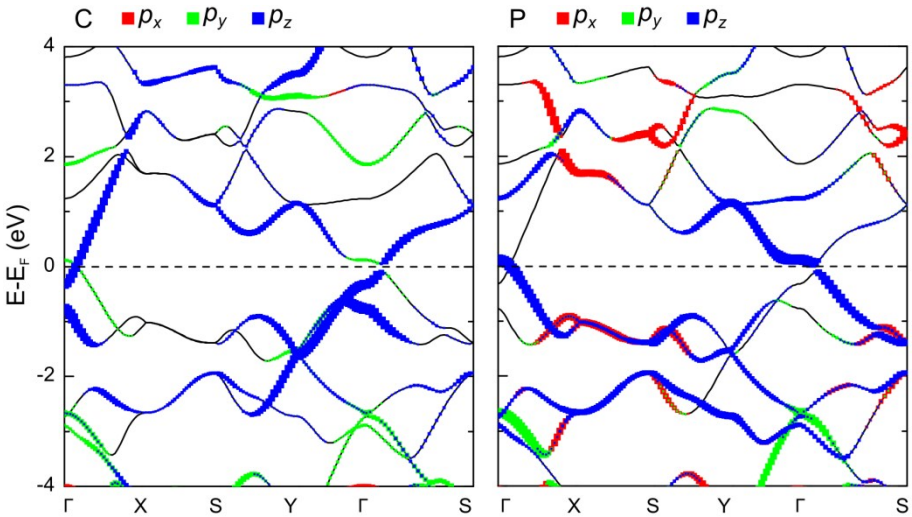

Figure S8. Atomic projected band structure associated with C atoms (left panel) and P atoms (right panel) for γ-CP monolayer.

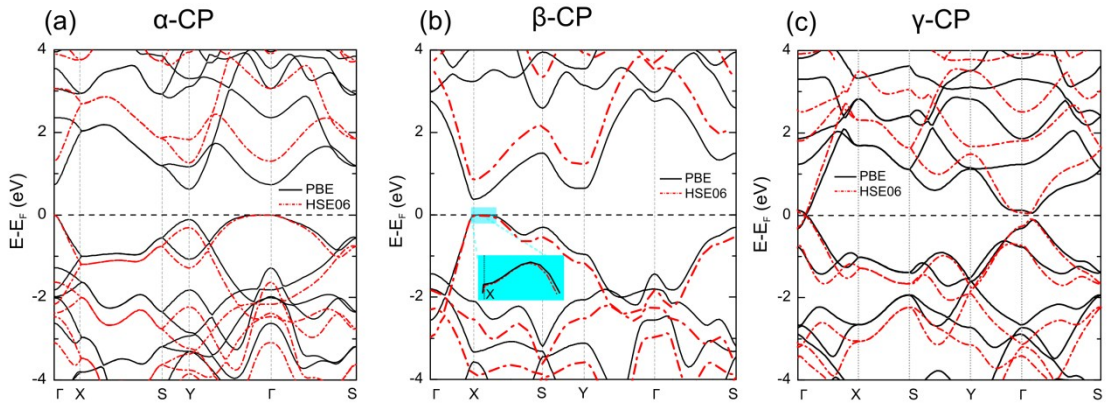

Figure S9. Comparison of the band structures obtained with GGA-PBE and HSE06 functional: (a) α-CP, (b) β-CP, and (c) γ-CP.

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