

# $\psi$ -Graphene: A New Metallic Allotrope of Planar Carbon with Potential Applications as Anode Materials for Lithium-Ion Batteries

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**Supporting Information** 

**ABSTRACT:** Using state-of-the-art first-principles calculations, we propose a new twodimensional (2D) carbon allotrope constructed by polymerizing the carbon skeletons of *s*indacenes, named PSI ( $\psi$ )-graphene. We show that  $\psi$ -graphene has the lowest energy among all hitherto reported 2D allotropes of carbon composed of 5–6–7 carbon rings and is dynamically and thermally stable. This structure is metallic with robust metallicity against external strain. In addition, we find that the adsorption of Li atoms on  $\psi$ -graphene is exothermic, and the diffusion energy barrier is low and comparable to that of graphene. Furthermore,  $\psi$ -graphene can achieve a maximum Li storage capacity equivalent to that of LiC<sub>6</sub>, suggesting its potential as an anode material for Li-ion batteries (LIBs). In addition, we show that increasing the number of hexagons in this structure can enhance the thermodynamic stability of the sheet while maintaining its metallicity. This study provides new insights into the design of new metallic carbon for nanostructured anode materials in the next generation of LIBs.



arbon, because of its ability to form sp,  $sp^2$ , and  $sp^3$ / hybridization, exhibits many allotropes. These include zero-dimensional (0D) C<sub>60</sub> fullerene,<sup>1</sup> one-dimensional (1D) carbon nanotubes<sup>2</sup> and nanoribbons,<sup>3</sup> 2D graphene,<sup>4</sup> and threedimensional (3D) graphite. Because of their different structures, these allotropes possess unique properties. Among these, graphene, the most stable 2D form of carbon, has attracted considerable attention due to its unusual electronic and spintronic properties, such as massless Dirac Fermions<sup>5</sup> and quantum Hall effect.<sup>6</sup> Moreover, the successful synthesis of graphene<sup>4</sup> has motivated scientists to explore other forms of 2D carbon. Because the geometric structure of graphene determines its unique properties,<sup>7,8</sup> the commonly used strategy for finding a new 2D carbon allotrope is to search for 2D carbon with hitherto unknown topology. A wealth of carbon allotropes composed of structural motifs such as tetragons, pentagons, heptagons, octagons, and so on have been theoretically predicted. Among these are pentaheptite,<sup>9</sup> haeckelites,<sup>10</sup> T graphene,<sup>8</sup> OPG,<sup>11</sup> pentahexoctite,<sup>12</sup> penta-graphene,<sup>13</sup> and phagraphene,<sup>14</sup> to name a few. Their electronic properties change from metal to semimetal and semiconductor, enriching potential applications of 2D carbon. Nevertheless, despite their outstanding properties, none of these allotropes has yet been synthesized due to experimental challenges. Therefore, it is desirable to explore new 2D carbon allotropes that are easier to synthesize.

In this regard, it is worth noting that direct assembly of their molecular precursors can lead to the synthesis of nanostructures with atomic-scale precision.<sup>15,16</sup> This bottom-up approach has been successfully employed in the synthesis of graphene sheet,<sup>17</sup> nanoribbons,<sup>18,19</sup> and nanotubes<sup>20</sup> as well as some other nanostructures.<sup>21–23</sup> With the assistance of suitable substrates, precursor molecules aggregate to form the extended system, whose topology is defined by the structure of the precursor monomers.

We use the above procedure to construct a 2D  $\psi$ -graphene sheet by regularly arranging the carbon skeletons of *s*-indacene, a small hydrocarbon molecule with a chemical formula C<sub>12</sub>H<sub>8</sub>. First-principles calculations combined with ab initio molecular dynamics simulations reveal that  $\psi$ -graphene is not only dynamically and thermally stable with its mechanical stiffness larger than that of penta-graphene<sup>13</sup> and *h*-BN monolayer<sup>24</sup> but also is metallic under ambient conditions. In addition, because of its unique structure and metallicity,  $\psi$ -graphene exhibits a low diffusion energy barrier and high storage capacity for Li ions. Thus  $\psi$ -graphene could be a promising anode material for LIBs. Furthermore, we extended the structure by increasing the number of the hexagons between the two neighboring 5–7 rings to tune the stability while maintaining its metallicity.

First-principles calculations based on density functional theory (DFT) are performed by using Vienna Ab initio

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Simulation Package (VASP).<sup>25</sup> Electron wave functions are expanded using the Projector Augmented Wave (PAW) method<sup>26</sup> with a kinetic energy cut off of 520 eV. Perdew-Burke-Ernzerhof (PBE)<sup>27</sup> functional with the Generalized Gradient Approximation  $(GGA)^{17}$  is used to treat the electron exchange-correlation interactions in most calculations, while Heyd–Scuseria–Ernzerhof (HSE06)<sup>28,29</sup> functional is used to predict more accurate electronic structure. The Brillouin zone is represented by k points with a grid density of  $2\pi \times 0.02$  Å<sup>-1</sup> in the reciprocal space using the Monkhorst-Pack scheme.<sup>30</sup> A vacuum space of 20 Å in the direction perpendicular to the sheet is used to separate it from its periodic images. The structure is relaxed without any symmetry constraint. The convergence threshold for total energy and atomic force components are  $10^{-5}$  eV and  $10^{-3}$  eV/Å, respectively. Phonon calculations are performed by using the finite displacement method<sup>31</sup> implemented in the Phonopy package.<sup>32</sup> Å (6 × 6) supercell is used to calculate the atomic force and the dynamical matrix. A tight-binding (TB) Hamiltonian is constructed based on the maximally localized Wannier functions (MLWFs). The hopping parameters are calculated by using the Wannier90 package<sup>33</sup> interfaced with the Quantum ESPRESSO code.<sup>34</sup> The climbing-image nudged elastic band (CI-NEB) method<sup>35,36</sup> is applied to determine the diffusion barriers of Li adatoms on the surface of  $\psi$ -graphene. To accurately search for the lowest energy adsorption configurations for  $LiC_x$  with varied x values, we use the particle-swarm optimization (PSO) algorithm implemented in the Crystal structure AnaLYsis by Particle Swarm Optimization (CALYPSO) code.<sup>37</sup> In the PSO method, we fix the coordinates of the C atoms in  $\psi$ -graphene, while allowing the coordinates of Li atoms to evolve, and the structure relaxations are performed using the DFT-based calculations.

Structure and Stability of  $\psi$ -Graphene. The optimized structure of  $\psi$ -graphene is plotted in Figure 1, which can be viewed as



Figure 1. Optimized structure of  $\psi$ -graphene with the *s*-indacene skeletons highlighted in yellow. Dashed rectangles represent the unit cell of  $\psi$ -graphene and C1~C4 are inequivalent carbon atoms in the unit cell.

parallel alignment of carbon skeletons of *s*-indacene, a hydrocarbon molecule composed of 5-6-5 carbon rings. Similar to graphene, the polymeric sheet is flat and one-atom thick. Because of the cyclization of the neighboring 5-6-5 carbon skeletons, the resulting 2D sheet is composed of 5-6-7 carbon rings. The optimized lattice constants of  $\psi$ -graphene are a = 6.70 Å and b = 4.84 Å, respectively, with 12 carbon atoms in one unit cell having *P2mg* plane symmetry (plane group no.

7). In this structure, there are four chemically inequivalent carbon atoms, labeled as  $C1\sim C4$  in Figure 1. The C1-C1 bond length (1.51 Å, highlighted by the light blue circle in Figure 1) is rather large compared with other bonds (1.41 to 1.44 Å) in the same structure. This is because this C1-C1 bond is at the meeting point of three *s*-indacene molecules.

To confirm the stability of this structure, we first compare the total energy of  $\psi$ -graphene with that of some other 2D carbon allotropes including graphene, haeckelite  $H_{567}^{10}$  and phagraphene.<sup>14</sup> The calculated total energies of  $\psi$ -graphene, graphene,  $H_{567}$ , and phagraphene in the equilibrium state are -9.07, -9.23, -8.99, and -9.03 eV/atom, respectively. The calculated dependence of the total energies on the area of these carbon allotropes is given in Figure 2a. This shows that although  $\psi$ -graphene is metastable compared with graphene, it is more stable than phagraphene and  $H_{567}$ .

Next, we calculate the phonons of  $\psi$ -graphene to study its dynamical stability. The results are presented in Figure 2b. The phonon spectra and density of states (DOS) show no imaginary modes in the entire Brillouin zone, confirming the dynamic stability of  $\psi$ -graphene. We also perform ab initio molecular dynamics (AIMD) simulations to examine the thermal stability of  $\psi$ -graphene. A (4 × 4) supercell containing 192 atoms is constructed to simulate the 2D sheet. From the results given in Figure S2, one can see that, after heating at temperature of 300 and 1000 K for 9 ps, the structure remains nearly intact without apparent distortion, and the total potential energy only fluctuates about a constant value, suggesting that the structure is thermally stable at room temperature and can even withstand temperatures as high as 1000 K.

Electronic Properties of  $\psi$ -Graphene. The electronic properties of  $\psi$ -graphene are investigated by calculating its electronic band structures and total DOS. The results are plotted in Figure 3a, which shows no gap between the valence and conduction bands and nonzero DOS at the Fermi level. The two partial occupied bands (labeled as I and II) cross the Fermi level, showing that  $\psi$ -graphene is metallic.

It is well known that the PBE functional underestimates the band gaps of semiconductors, and a semiconductor with a very small band gap may be erroneously concluded to be a metal. To improve accuracy, we calculate the band structures of  $\psi$ -graphene by using HSE06 hybrid functional; the results are plotted in Figure 3a. We see that  $\psi$ -graphene remains metallic.

We have also examined the dependence of electronic property on external strain. By stretching or compressing the structure along different directions (*a*-axial, *b*-axial, and equibiaxial) up to 6%, we find that  $\psi$ -graphene remains metallic regardless of the direction and magnitude of strain (see Figure S3).

The electronic states near the Fermi level can be further mapped by the Fermi surface of  $\psi$ -graphene, which is shown in Figure 3b. Purple and blue circles represent electron and hole pockets, respectively. It shows that in the first Brillouin zone there are one electron pocket and two hole pockets. The electron pocket is centered around X point and originated from band II, while the hole pocket is located on the X–M path and originated from band I. Band I and band II are entangled along the X–M path, and thus the electron and hole pockets make contact in the momentum space, with each electron pocket sandwiched by two hole pockets. However, because the crossing point is not located at the Fermi energy, the neighboring electron and hole pockets in the Fermi surface do not touch.



Figure 2. (a) Area dependence of total energies for some 2D carbon allotropes. (b) Phonon band structures and DOS of  $\psi$ -graphene.



**Figure 3.** (a) Electronic band structures and DOS (the red/blue lines represent the HSE06/PBE results) and (b) Fermi surface of  $\psi$ -graphene. ELF of  $\psi$ -graphene with slices (c) crossing the structure plane and (d) 1 Å above the structure plane.



Figure 4. (a) Orbital decomposed band structures of  $\psi$ -graphene. (b) Band structures derived from the TB model Hamiltonian and DFT (PBE).

To further explore the bonding feature and origin of metallicity of  $\psi$ -graphene, we calculate the electron localization

function (ELF), which is widely used to analyze the spatial distribution of electrons. Values of ELF is renormalized

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**Figure 5.** (a) Considered Li adsorption sites on the surface of  $\psi$ -graphene. Blue, green, and red dots represent the hollow, top, and bridge sites, respectively. (b) The four possible migration paths and (c) the corresponding diffusion energy profiles of Li diffusion on  $\psi$ -graphene.

between 0.0 and 1.0. The values 0.5 and 1.0 represent fully delocalized (homogeneous electron gas) and fully localized electrons, while the value 0.0 refers to a very low charge density. From the ELF slice presented in Figure 3c, one can see that the electrons are localized on the C–C bonds, indicating the strong  $\sigma$  bonding states between the neighboring C atoms. However, when the slice is moved 1 Å up from the plane of the structure (Figure 3d), the electrons distribute uniformly and form a delocalized electron network (ELF  $\approx$  0.5). This delocalized electron network forms the metallic channels, which results in the metallicity of  $\psi$ -graphene.

To determine the orbital composition of the metallic states, we calculate the orbital decomposed band structures of  $\psi$ graphene. The results are shown in Figure 4a. One can see that the electronic states near the Fermi level originate solely from the  $p_z$  orbitals, while the s,  $p_{xx}$  and  $p_y$  orbitals hybridize together forming the deep-lying energy states. This is consistent with the ELF results: (1) the deep-lying energy states correspond to the low-energy  $\sigma$ -bonding states, which are formed by the hybridization of s,  $p_{xy}$  and  $p_y$  orbitals; (2) the  $p_z$  orbitals conjugate to form the delocalized  $\pi$  bonds, and the corresponding  $\pi$  bands cross the Fermi level, giving rise to metallicity of the structure. Moreover, we note that the bands with  $p_z$ -orbital characteristics are entirely decoupled from the others. Therefore, the dispersions of these  $\pi$  bands can be described by using a simplified TB Hamiltonian of C- $p_z$  orbitals

$$H = U \sum_{i} c_i^{\dagger} c_i + t \sum_{\langle i,j \rangle} c_i^{\dagger} c_j$$
<sup>(1)</sup>

Here  $c_i^+$  and  $c_i$  represent the creation and annihilation operators of an electron at site *i*, *U* is the onsite energy, and *t* is the hopping integral parameter between nearest neighbor *i* and *j* sites. By fitting the MLWF-based TB band structures with the DFT results, the onsite energy *U* and the hopping integral parameter *t* are determined to be 0.63 and -2.80 eV, respectively. After diagonalizing the TB model Hamiltonian, we obtain the electronic bands of  $\psi$ -graphene, which are plotted in Figure 4b. It shows that by only considering the nearest neighbor hopping, the TB bands can reproduce well the alignment and dispersion features obtained from the DFT results. In addition, the nearest-neighbor hopping integral in  $\psi$ -graphene is similar to that in graphene (-2.70 eV<sup>11</sup>), from which we deduce that the difference of electronic structures between  $\psi$ -graphene and graphene results from their different atomic structures.

Mechanical Properties of  $\psi$ -Graphene. In addition to the electronic properties, we also investigate the mechanical properties of  $\psi$ -graphene by using the energy-strain method, which has been successfully employed in many 2D materials.<sup>13,38–40</sup> In a 2D sheet, the elastic strain energy per unit area can be expressed as<sup>13</sup>

$$U(\varepsilon) = \frac{1}{2}C_{11}\varepsilon_{xx}^{2} + \frac{1}{2}C_{22}\varepsilon_{yy}^{2} + C_{12}\varepsilon_{xx}\varepsilon_{yy} + 2C_{66}\varepsilon_{xy}^{2}$$
(2)

By fitting the strain-energy curves associated with uniaxial and equi-biaxial strain (Figure S4), we obtain the elastic constants  $C_{11}$ ,  $C_{22}$ , and  $C_{12}$  to be 306.73, 320.42, and 84.44 N/m, respectively. The in-plane Young's modulus along a ( $E_a$ ) and b ( $E_b$ ) directions, which can be derived from the elastic constants by using equations  $E_a = (C_{11}^2 - C_{12}^2)/C_{11}$  and  $E_b = (C_{22}^2 - C_{12}^2)/C_{22}$ , are calculated to be 283.49 and 298.17 N/m, respectively. The different values of  $E_a$  and  $E_b$  indicate that  $\psi$ -graphene is mechanically anisotropic. From the elastic constants, we can also deduce the Poisson's ratio using  $\nu_a = C_{12}/C_{11}$  and  $\nu_b = C_{12}/C_{22}$ . The calculated  $\nu_a$  and  $\nu_b$  are 0.275 and 0.264, respectively. Although the Young's moduli of  $\psi$ -graphene are smaller than those of graphene ( $E_a = E_b = 342.2 \text{ N/m}^{24}$ ), they are larger than those of penta-graphene ( $E_a = E_b = 263.8 \text{ N/m}^{13}$ ) and h-BN monolayer ( $E_a = E_b = 275.8 \text{ N/m}^{24}$ ), suggesting that  $\psi$ -graphene has good mechanical properties.

Potential Application of  $\psi$ -Graphene as Anode Materials for Li-Ion Batteries. Because of its outstanding mechanical property and electronic conductivity<sup>41</sup> as well as high surface-to-mass ratio,<sup>42</sup> graphene has been intensively studied both experimentally<sup>43-45</sup> and theoretically<sup>46-48</sup> as an alternative anode material for LIBs. However, on the basis of DFT calculations combined with the cluster expansion method, it was found that Li atoms do not absorb on defect-free single-layer graphene,



**Figure 6.** (a) Structure illustration of *n*-hex- $\psi$ -graphene. Red rectangle represents the unit cell of the structure. (b) Energy dependence on *n* of *n*-hex- $\psi$ -graphene. (c) Electronic band structures of *n*-hex- $\psi$ -graphene. The left panel: 3-hex- $\psi$ -graphene; the right panel: 13-hex- $\psi$ -graphene. (d) Band decomposed charge density of 13-hex- $\psi$ -graphene (blue: the 120th band; green: the 121th band).

which hinders its applications as an electrode material.<sup>49</sup> Several investigations were performed to explore the feasibility of improving Li capacity by modifying graphene.<sup>50–52</sup> It was found that monovacancy, divacancy, and Stone–Wales defects can create active sites for Li adsorption.<sup>53,54</sup> We expect that  $\psi$ -graphene with its intrinsic 5–7 carbon rings would not require additional defects for Li adsorption.

To examine this possibility we first calculate the adsorption energy of a single Li atom on a  $2 \times 2$  supercell of  $\psi$ -graphene. We define this energy as

$$E_{\rm a} = E_{\psi + \rm Li} - E_{\psi} - E_{\rm Li} \tag{3}$$

where  $E_a$  is the adsorption energy,  $E_{w+Li}$  and  $E_w$  are the total energies of  $\psi$ -graphene supercell after and before Li adsorption, and  $E_{Li}$  represents the energy per Li atom in bulk Li metal. Because the weak van der Waals (vdW) interactions are less significant in single-layer systems,<sup>54</sup> we do not include the vdW correction in our calculations. Because of the P2mg plane symmetry of  $\psi$ -graphene, there are 14 possible adsorption sites, namely, four hollow sites (H1~H4), four top sites (T1~T4), and six bridge sites (B1~B6), as shown in Figure 5a. After structure optimization, we find Li atoms to preferably adsorb on the center of the carbon rings, consistent with previous studies for Li adsorption on graphene surface with or without defects.<sup>54</sup> The calculated adsorption energies for H1, H2 (H3 site has the same  $E_a$  as H2 site), and H4 sites are -0.64, -0.58, and -0.62 eV, respectively, comparable to that of the widely used anode material, graphite (-0.78 eV) and recently predicted bco- $C_{16}$  (-0.63 eV).<sup>55</sup> In contrast with the case of graphene with low density of defects, the adsorption energies for different hollow sites are very similar, which can be attributed to the high density of 5–7 rings of  $\psi$ -graphene. The negative adsorption energies indicate that Li atoms will not cluster and preferably bind at the H1 site. Furthermore, the

Bader charge analysis<sup>56</sup> shows that the charge transfer from Li to the  $\psi$ -graphene sheet is ~0.89 lel for all of the cases.

The potential of  $\psi$ -graphene as an anode material in LIBs also depends on the mobility of the Li ions. We study the diffusion of Li adatoms on the surface of  $\psi$ -graphene by using the CI-NEB method. We consider the migration pathways of Li adatoms from its preferred adsorption site H1 to the nearest-neighbor H1 site along the crystal axes *a* and *b*. The four possible migration pathways and the corresponding diffusion energy profiles are presented in Figure 5b,c, respectively. We see that the diffusion barriers of the two paths along the *a* axis are both 0.313 eV, while for the two paths along the *b* axis the diffusion barriers are 0.295 and 0.265 eV, respectively. These barriers are comparable to that of graphene (0.31 eV).<sup>57</sup> The high Li-ion mobility indicates good charge–discharge rates when  $\psi$ -graphene is used as an anode for LIBs.

In practice, the storage capacity of batteries is a key indicator of the performance of electrode materials. To this end we increase the concentration of Li adatoms and explore the maximum capacity of Li on  $\psi$ -graphene. Lowest energy adsorption configurations of  $LiC_x$  with different x values as well as the adsorption energies for these configurations are calculated by using the PSO method. The results are given in the SI. Confining the adsorption of Li to only one side of  $\psi$ graphene, the maximum theoretical specific capacity can reach 372 mA h g<sup>-1</sup>, the same as that of graphite,<sup>58</sup> but much larger than that in pristine graphene. In this LiC<sub>6</sub> stoichiometry, the lowest energy configuration is that where two Li adatoms are absorbed on two H4 sites per unit cell of  $\psi$ -graphene. Although the adsorption energy of -0.01 eV/atom is low and indicates weak binding, it is comparable to or even larger than that of several typical electrode materials, such as Li on Mo<sub>2</sub>C (adsorption energy -0.01 eV/atom)<sup>59</sup> and Na on Ca<sub>2</sub>N (adsorption energy -0.003 eV/atom).<sup>60</sup> Next, we estimate the

average open-circuit voltage  $(V_{\rm ave})$  by using the following equation

$$V_{\text{ave}} = (E_{\psi} + xE_{\text{Li}} - E_{\text{Li}_x - \psi})/xe \tag{4}$$

The calculated  $V_{ave}$  is 0.64 V for a stoichiometry of LiC<sub>48</sub>. As the adsorbed concentration increases to LiC<sub>6</sub>,  $V_{ave}$  decreases to 0.01 V. Low-average open-circuit voltage is beneficial to the overall voltage of the cell, which means that once the  $\psi$ graphene anode is connected to the cathode, the LIB can supply a higher operating voltage with larger energy capacity. The exothermic adsorption, low diffusion energy barrier, and high specific capacity of Li adatoms as well as the robust intrinsic metallicity and good mechanical strength imply that  $\psi$ graphene can be a promising anode material for LIBs.

Extended Structures of  $\psi$ -Graphene. The structure of  $\psi$ graphene can be viewed as a disordered graphene with parallelaligned 5-7 line defects separated by hexagons. By increasing the ratio of hexagons in the structure, we generate a series of extended structures. The hexagon-rich carbon structure is termed n-hex- $\psi$ -graphene, where n refers to the number of hexagons between the two neighboring 5-7 rings along the crystal axis  $a_i$  as illustrated in Figure 6a. As the value of nincreases, the energy of the structure decreases and finally approaches that of pristine graphene (-9.23 eV/atom, see)Figure 6b). The electronic band structures of the extended structures are calculated at the PBE level. The results are presented in Figure 6c and Figure S7. One can see that for different n values, the structures are all metallic as the two partially occupied bands, namely, the two bands crossing the Fermi level and contributing to the metallicity of the structure, remain nearly unchanged. To understand the robust metallicity with respect to n, we calculate the band decomposed charge density of 13-hex- $\psi$ -graphene, where the distance between the two neighboring 5-7 rings along the crystal axis *a* is 2.71 nm. With this large distance, we can view this structure as graphene sheet with 1D 5-7 line defects. From the results shown in Figure 6d, we find that the main contribution to charge density comes from the carbon atoms of 5-7 rings, whose electrons of p<sub>z</sub> orbitals form the delocalized metallic wires along the 1D line defects, resembling that of 1D 5-5-8 topological line defects in graphene.<sup>61</sup> Therefore, *n*-hex-*ψ*-graphene will retain its metallicity regardless of the n value, which makes it convenient for practical applications.

In summary, using first-principles calculations based on DFT, we propose a new metallic allotrope of 2D carbon,  $\psi$ -graphene, which can be synthesized by using s-indacene derivatives as molecule precursors. This structure composed of 5-6-7 carbon rings is the lowest energy allotrope of planar carbon and displays the following properties. (1) It is dynamically and thermally stable. (2)  $\psi$ -Graphene is intrinsically metallic, and the structure can be flexibly engineered via incorporating more hexagons without compromising its metallicity. (3)  $\psi$ -Graphene has a fairly high in-plane stiffness comparable to that of graphene, preventing possible severe volume expansion upon Li adsorption, as is the case with silicon-based anodes. The metallicity of  $\psi$ -graphene is robust against external strain, and hence the electrical conductivity is not influenced even if the structure endures thermal or chemically induced strain. (4) The metallic  $\psi$ -graphene contains active sites for Li adsorption with a high specific capacity (372 mA h  $g^{-1}$ ) and a lower diffusion energy barriers for Li ions (<0.313 eV). Besides its potential as an anode material, metallic  $\psi$ -graphene may also have applications in catalysis, superconductivity, and electronic

devices. We hope that the present study will stimulate experimental efforts in synthesizing this new metallic 2D carbon, thus expanding the carbon family with potential for technological applications.

## ASSOCIATED CONTENT

#### **S** Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.jpclett.7b01364.

Geometric structures of some other 2D carbon allotropes; molecular dynamics simulations; electronic band structures under external strain; strain-energy curves; storage capacity of Li ions; electronic band structures of the extended structures; and possible experimental realization. (PDF)

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#### Notes

The authors declare no competing financial interest.

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