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Synthesis of new silicene structure and its energy band properties*

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Silicene, silicon analogue to graphene which possesses a two-dimensional (2D) hexagonal lattice, has attracted increasing attention in the last few years due to predicted unique properties. However, silicon naturally possesses a three-dimensional (3D) diamond structure, so there seems to be not any natural solid phase of silicon similar to graphite. Here we report the synthesis of new silicene structure with a unique rectangular lattice by using a coherent electron beam to irradiate amorphous silicon nanofilm produced by pulsed laser deposition (PLD). Under the irradiation of coherent electron beam with proper kinetic energy, the surface layer of silicon nanofilm can be crystallized into silicene. The dynamic stability and the energy band properties of this new silicene structure are investigated by using first-principle calculations and density function theory (DFT) with the help of the observed crystalline structure and lattice constant. The new silicene structure has a real direct bandgap of 0.78 eV. Interestingly, the simulating calculation shows that the convex bond angle is 118° in the new silicene structure with rectangular lattices. The DFT simulations reveal that this new silicene structure has a Dirac-cone-like energy band. The experimental realization of silicene and the theoretically predicted properties shed light on the silicon material with potential applications in new devices.

Keywords: silicene, nanofilm, irradiation of coherent electron beam, pulsed laser deposition, Dirac cone

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1. Introduction

Graphene has received the worldwide attention since the first experimental preparation was reported in 2004 due to its fascinating properties and many device applications. Novoselov, *et al*. discovered the electric field effect in thin carbon films and the two-dimensional gas of massless Dirac fermions in grapheme.[1,2] Kim *et al*. investigated the growth of graphene films for stretchable transparent electrodes.[3] Seol *et al*. studied the phonon transport in supported grapheme in 2010.[4] Since then, two-dimensional (2D) materials have gained rapid development. In the last few years, Feng *et al*.[5] and Lalmi, *et al*.[6] have dedicated their strong effort to growing 2D silicon material, which was expected to have a great impact on the development of future electronic devices and energy storage.[7–9] The 2D silicon material was named either analogue of graphene in Takeda and Shiraishi's work (1994)[10] or silicene in Guzmán-Verri and Lew Yan Voon's work (2007).[11] Recent relevant theoretical work done by Tsai *et al*.,[12] Tritsaris *et al*.,[13] and Tao, *et al*.[14] revealed that the silicene has a structure of Si atoms packed in a buckled honeycomb lattice, and it possesses massless Dirac fermions and other attractive properties,[12–14] which Liu *et al*.,[15] Vogt *et al*.,[16] and Linghu, *et al*.[17] have also made stronger effort to investigate . However, the valence electrons in silicon are localized in σ bonds and less mobile than those in graphite, which, as indicated by Yamada-Takamura and Friedlein,[18] can prevent the extended π electronic states from forming at low binding energy.[18] Therefore, the silicon crystal naturally possesses the three-dimensional (3D) diamond structure and no natural solid phase of 2D silicon crystal. The silicene really belongs to the kind of quasi-2D silicon crystal. Typically, Lin *et al*.,[19] Jamgotchian *et al*.,[20] and Scalise, *et al*.[21] prepared silicene on metal substrates by depositing silicon. Other structures such as silicene nanoribbons have also been prepared on Ag (110), in which graphene-like electronic signature was observed by Aufray *et al*.[22] and De Padova, *et al*.[23] The electronic properties of silicene grown on Ag (111) were also investigated by Chiappe, *et al*.[24] Although silicene has been demonstrated to have unique properties, their synthesis on silicon directly is still difficult to implement. The realization of such a structure would be able to fully explore its potential.

In addition, Morishita *et al*.[25] and Nishio, *et al*.[26] conducted the molecular dynamics simulation to study the formation of nanowire and double-layer silicon in slit pores. Their stability was further confirmed by first principles calculation.

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The calculations demonstrated the possibility of the synthesis of novel nanosilicon by confinement in nanopores. And Bai, *et al*. suggested that the bilayer hexagonal silicon is a quasi-2D semimetal in the double-layer silicon computation.[27] It is interesting to make a comparison among various 2D lattices in quasi-2D silicon crystals suggested in the previous studies, such as the two pairs of electromagnetically induced atomic gratings provided by Zhang *et al*.,[28] the 2D atomic lattices prepared by Zhang *et al*.,[29] and the optically induced atomic lattices in atomic media completed by Zhang *et al*.[30] Recently, two new 2D silicon allotropes (Si-*Cmma* and Si-*Pmma*) with excellent energetic stability and positive dynamical stability have been proposed and investigated by first-principles calculations in the work of Zhou, *et al*.[31]

It may be considered as another route to prepare new 2D structures of silicon crystal by using some novel methods. In the present article, we fabricate the quasi-2D structures of silicon crystal involving two kinds of silicene structures with the hexagonal lattice and the new rectangular lattice by using the coherent electron beam to irradiate the amorphous Si film produced through using the pulsed laser deposition (PLD) method, where quasi-2D crystallizing occurs due to suitably transferring energy of high speed electrons in layer, originating from the Heisenberg principle related to $\Delta p \sim h/\Delta x$. We investigate the dynamic stability of the new quasi-2D silicon structure by using first-principle calculations, which has a real direct bandgap of 0.78 eV near X region. We make a comparison between optimum quasi-2D structures of silicon crystal and ideal model of 2D structures of silicon crystal involving two kinds of the silicene structures with the hexagonal lattice and the new rectangular lattice in the simulating calculation. Total energy calculations by using density functional theory (DFT) show that the energy band in the new quasi-2D silicon structure possesses a Dirac-cone-like shape. Our experimental and theoretical study expand the new quasi-2D silicon structures and demonstrate the potential for new materials with desirable properties.

2. Method

Here, we report a new method of directly synthesizing silicene. It has been discovered in our experiment that the silicene crystal rapidly grows with coherent electron beam irradiating the amorphous silicon film prepared by using the PLD method on substrate. It is very interesting that the electronic irradiation promotes the growth of nanocrystal in silicene structure, whose physical mechanism may originate from the nanoscale characteristics of electronic de Broglie wave which produces resonance to transfer energy to crystal atoms, where the electron speed is very high due to the Heisenberg principle related to $\Delta mv \sim h/\Delta x$ in nanolayer. As is well known, in natural sciences, many analogous structures and properties occur in hierarchies with different sizes, such as on a sub-micrometer scale related to photonic de Broglie wavelength. The nanosecond or femtosecond laser was used by Shi *et al*.[32] to fabricate amorphous silicon film on a scale of hundreds of nanometers; and in the nanoscale space related to electronic de Broglie wavelength, the coherent electron irradiation was used by Huang, *et al*.[33] to prepare the silicon crystal structures on an angstrom scale.[33]

A silicon wafer (100) oriented substrate was placed on the sample stage in the PLD fabrication system. A pulsed Nd:YAG laser (wavelength: 1064 nm, pulse length: 60 ns, repetition rate: 1000) was used in PLD process. Figure 1 shows the synthesis process of the silicene, which involves a novel two-step fabricating process: the first step is to produce the amorphous silicon film by using the PLD method, and the second step is to generate the silicene crystalline by using the coherent electron beam to irradiate the film. In the second process. The density of electron beam was about 0.5 nA/nm^2. The electron beam was produced from field-emission electron gun in the Tecnai G2 F20 system and accelerated by 200 kV to higher energy, and it was better coherent. Under the irradiation of coherent electron beam, the silicene crystal was produced by controlling irradiating time from 20 min to 30 min.

3. Result

The PLD process is shown in Fig. 1(a), a nanosecond pulsed laser irradiates the silicon surface to form plasma consisting of silicon ions and electrons, and its interaction with silicon leads the amorphous silicon film on a nanoscale to be deposited on the silicon surface. Figure 1(b) shows the cross sectional and top-view TEM images of the amorphous silicon layer, which is prepared on an amorphous carbon substrate. Figure 1(c) shows the detailed growth process of the silicene crystal occurring under the coherent electron beam irradiating the amorphous silicon film in an Ar atmosphere of 10^{-7} Pa. In Fig. 1(d), on the left are the TEM images of spot shape under the coherent electron beam irradiating the surface of the amorphous silicon film, and on the right is the TEM image of the silicene crystal growing under the interaction of coherent electron beam with surface.

It is important that besides the silicene with the hexagonal lattice that is observed in the TEM image of Fig. 2(a), the new silicene crystal with rectangular lattice be also discovered under the 20–30-min irradiation of coherent beam on the amorphous silicon nanofilm produced by using the PLD method as shown in the TEM image of Fig. 2(b) with the inset exhibiting the electronic diffraction pattern on the 2D rectangular lattice. Here, the analysis of TEM image exhibits that the new silicene structure with rectangular lattice occurs in monoatomic layer on amorphous silicon. Here, the lattice constant is about 0.23 nm, and the electronic diffraction pattern is related to the rectangular lattice.

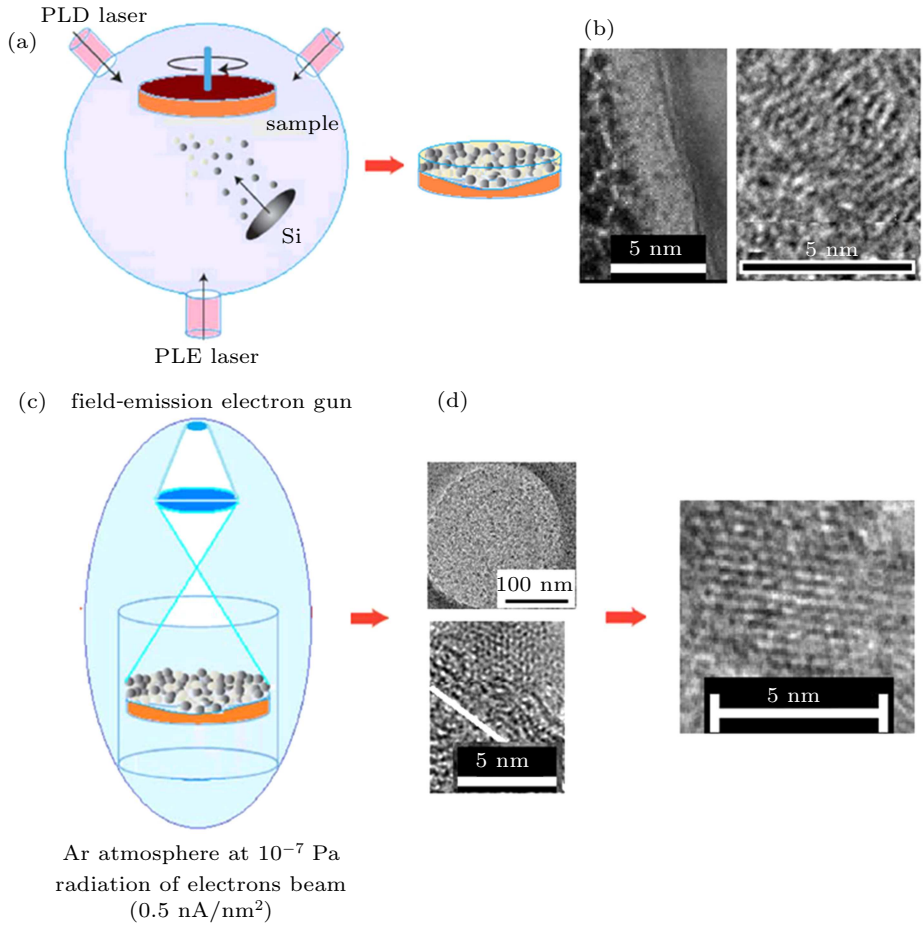


Fig. 1. (a) Fabrication process of amorphous silicon nanofilm by using PLD method, (b) TEM images with nanofilm structure of amorphous silicon, (c) preparing process of silicene crystal by using coherent electron beam to irradiate amorphous silicon nanofilm, and (d) TEM image of spot shape occurring with coherent electron beam irradiating surface (left), and TEM image of the silicene crystal (right).

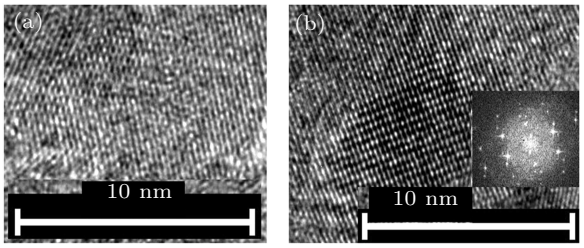


Fig. 2. (a) TEM image of silicene with hexagonal lattices, (b) TEM image of new silicene with rectangular lattices, with inset exhibiting electronic diffraction pattern of rectangular lattice.

The dynamic stability of the silicene is investigated by using first-principles calculations with consideration of two kinds of crystal structures observed in experiment, *i.e.*, the hexagonal lattice and the new rectangular lattice. Their electronic behavior is investigated by an ab initio nonrelativistic quantum mechanical analysis. The density functional theory (DFT) is used to calculate the density of states (DOS) on Si 2D structure, which is carried out with the local density approximation (LDA) and gradient-corrected exchange–correlation function (GGA) for the self-consistent total energy method. It is interesting to note that in the total energy calculations using DFT, the quasi-2D structure of silicon crystal with the convex bonds occurs at the lowest energy of optimum structure through the optimizing process of the ideal primal model of 2D silicon crystals.

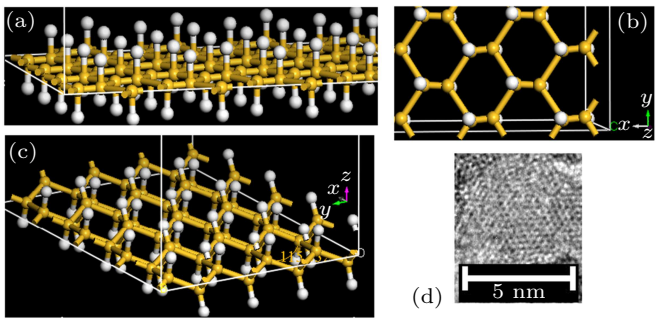


Fig. 3. Simulation model of ideal primal 2D silicon with hexagonal lattice passivated by hydrogen atoms (white balls), showing (a) laterally cross-cut structure of ideal primal model in 2D structure with hexagonal lattice, (b) overlook structure of ideal primal model in 2D structure with hexagonal lattice, (c) silicene structure with hexagonal lattice after optimizing process in simulating calculation, in which convex bond angle of silicon atom is about 115°, and (d) TEM image of silicene crystal with hexagonal lattice observed in experiment.

At first, for an ideal primal model of 2D silicon crystal with a hexagonal lattice, its cross sectional structure is shown in Fig. 3(a) and its top view before optimizing process in the simulating calculation is displayed in Fig. 3(b), where the surface of the ideal primal 2D structure needs to be passivated by

hydrogen atoms (white balls) and has a simple and effective treatment of surface dangling bonds in simulation. It should be noted that after optimizing process, the quasi-2D structure with convex bonds is formed in a hexagonal lattice as shown in Fig. 3(c), in which the convex bond angle of silicon atom on the quasi-2D is about 115°. Here, it is interesting to make a comparison between the silicene structure produced in the optimized process of calculation as shown in Fig. 3(c) and the silicene with the hexagonal lattice in the TEM image observed in the experiment as exhibited in Fig. 3(d), showing that they are very likeeach other in the lattice shape.

According to the simulation model of the silicene crystal with the hexagonal lattice related to the experimental result, the energy band is calculated to have a direct bandgap of 1.4 eV as shown in Fig. 4(a), where a nearly Dirac-cone-like shape occurs at Γ point. The density of electronic states is calculated as shown in Fig. 4(b), in which the localized states occur in bandgap.

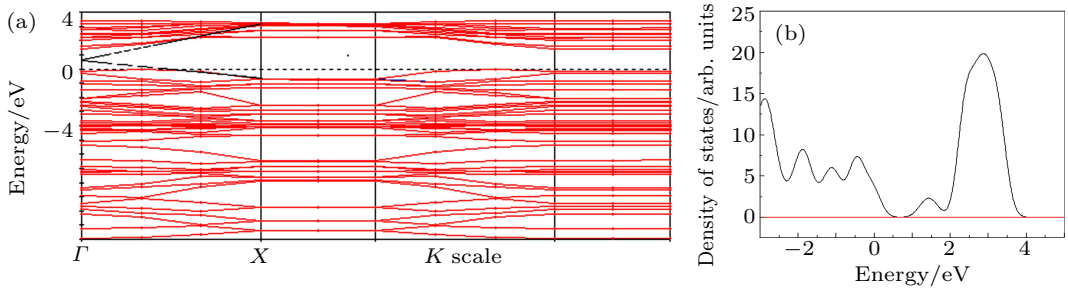


Fig. 4. (a) Energy band structure calculated in simulation model of silicene with hexagonal lattice, where it has a direct bandgap of 1.4 eV and a nearly Dirac-cone-like shape at Γ point, (b) density distribution of states with localized peak near conduction band valley calculated in simulation model of silicene with hexagonal lattice.

And then, we come to investigate the new silicene structure with the rectangular lattice observed in experiment by using first-principles calculations.

The ideal primal model of 2D silicon crystal with the rectangular lattice in simulation is built as shown in Fig. 5. The lateral crosscut and the overlook of ideal primal 2D rectangular lattice are exhibited in Figs. 5(a) and 5(b), respectively. The rectangular lattice of the 2D structure of silicon crystal has a four-fold symmetry. The lattice points of a 2D Bravais lattice assumed to be in the xy plane, are given by the vectors: $X(l) = l_1\boldsymbol{a}_1 + l_2\boldsymbol{a}_2$, where $\boldsymbol{a}_1$ and $\boldsymbol{a}_2$ are the primitive translation vectors of the lattice. And the diffraction points are described by $G(h) = h_1\boldsymbol{b}_1 + h_2\boldsymbol{b}_2$, where $\boldsymbol{b}_1$ and $\boldsymbol{b}_2$, which are the primitive translation vectors of the reciprocal lattice. In the 2D Bravais lattices, the structure parameters for the rectangular lattice of the new 2D structure of silicon crystal are as follows: $\boldsymbol{a}_1$ $(a_0, 0)$, $\boldsymbol{a}_2$ $(0, a_0)$, $\boldsymbol{b}_1$ $(2\pi/a_0(1,0))$, $\boldsymbol{b}_2$ $(2\pi/a_0(0,1))$, and a_c (a_0^2).

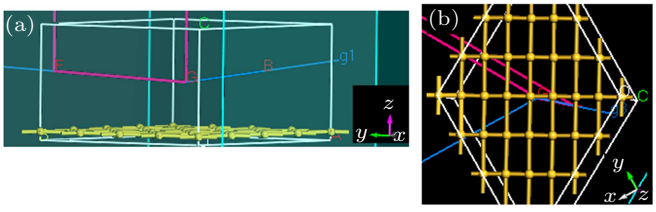


Fig. 5. Simulation model of 2D crystal with rectangular lattice, showing (a) laterally cross-cut structure of ideal primal model of 2D structure of silicon crystal with rectangular lattice, and (b) overlook structure of ideal primal model of 2D structure of silicon crystal with rectangular lattice.

The interaction energy for the rectangular lattice of 2D silicon crystal in simulation can be optimized to keep lower, thus making this lattice more stable. In the geometry optimization, figure 6(a) shows the change process of structure energy which is lower than that with the hexagonal lattice in the simulating calculation, where the structure parameters can be obtained through the optimization process in the DMol3 mode and the CASTEP mode. And in Fig. 6(b), the simulating result shows the new silicene structure with the rectangular lattice after optimizing process, the convex bond angle of silicon atom of about 118°, the lattice constant of about 0.23 nm and the thickness of atomic layer of about 0.12 nm. The structure, nearly monoatomic layer, possesses a fractional dimension of 2.1–2.5 in the quasi-2D case, and transforms into an ideal quantum-film shape, where new quantum phenomena and effects will appear in the process from the quasi-2D shape to the idea quantum-film.

The new silicene crystal with the rectangular lattice is synthesized with the interaction of electron beam with surface of amorphous silicon nanofilm produced by PLD method. Here, it is interesting to make a comparison between the new silicene structure in optimized calculation and the new silicene with the rectangular lattice in the TEM image observed in experiment as exhibited in Fig. 6(c). The new silicene crystal with the rectangular lattice is more stable, which was demonstrated experimentally for several months and is displayed in the present simulating calculation for the lowest binding energy of the structure.

According to the simulation model of the new silicene crystal with the rectangular lattice related to the experimental result, the energy band structure is calculated and the results are exhibited in Fig. 7(a), showing a real direct bandgap of 0.78 eV, and a nearly Dirac-cone-like shape occurring near the X region. Here, the different bandgap from that in the silicene

crystal with the hexagonal lattice may be due to the difference in combining energy. The density of electronic states is calculated as displayed in Fig. 7(b), showing the localized peaks near the Fermi level and conduction band valley occurring in the bandgap. Here, the direct bandgap of 0.78 eV (corresponding to a wavelength of about 1500 nm) in the new silicene crystal should have a good application in optic-communication window.

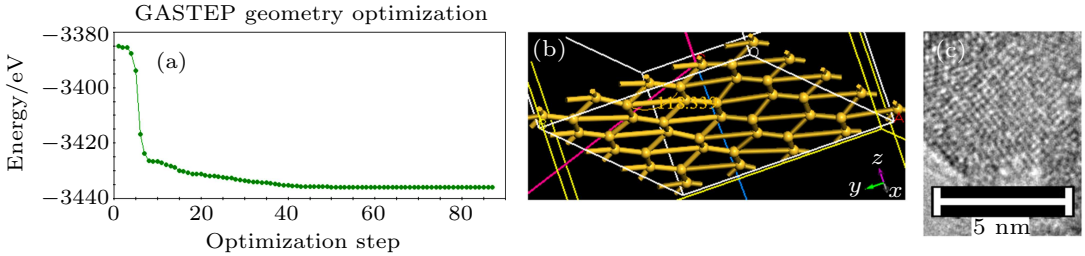


Fig. 6. Change process in CASTEP geometry optimization of new silicene crystal with rectangular lattice, showing (a) change process of structure energy in optimization process, (b) new silicene crystal with rectangular lattice after optimizing process in simulating calculation, in which convex bond angle of silicon atom is about 118°, and (c) TEM image of new silicene crystal with rectangular lattice observed in experiment.

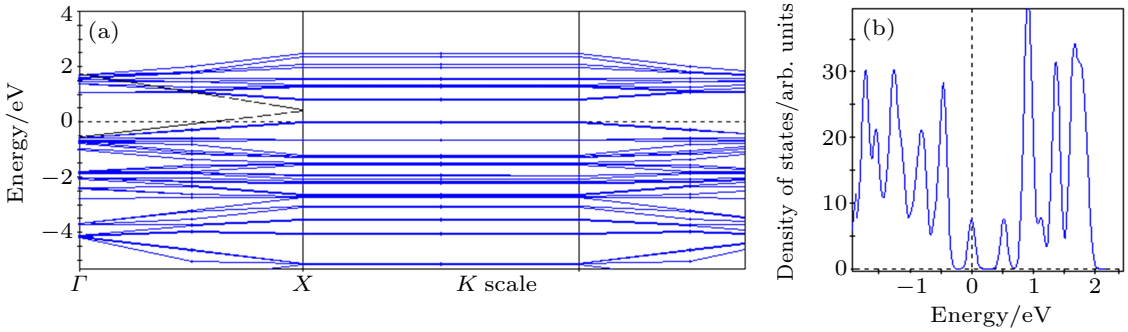


Fig. 7. (a) Calculated energy band structure for new silicene crystal with rectangular lattice showing a real direct bandgap of 0.78 eV and a nearly Dirac-cone-like shape in X region, and (b) simulated density distribution of states for new silicene crystal with rectangular lattice, showing localized peaks in bandgap.

4. Conclusion

In this work, we present a new method to synthesize the silicene directly by using a coherent electron beam to irradiate an amorphous silicon film prepared by the pulsed laser deposition. The prepared silicene structure exhibits a new rectangular lattice observed in experiment, where the rectangular lattice structure remains at the lowest binding energy demonstrated in simulating calculation. The energy band properties are simulated by using first-principles calculations. The simulating calculation shows a direct bandgap of 0.78 eV (corresponding to a wavelength of about 1500 nm) in the new silicene crystal with the rectangular lattice, and the direct bandgap should have a good application in optic-communication window. The total energy calculations by using DFT reveal that the new silicene structure has a nearly Dirac-cone-like energy band of graphene. It is interesting to observe the structure changing between the silicene crystal after being optimized and the ideal primal model of 2D structure of silicon crystal in the simulating calculation, in which the convex atomic layers are found in the quasi-2D structure that is optimized by replacing the plat atomic layer in the ideal primal model. The discovery of new silicene structures prepared on silicon directly and their theoretically predicted unique properties shed light on conventional silicon material and open a new route towards the era of nanoelectronic silicon devices.

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