Hexagonally Warped Dirac Cones and Topological Phase Transition in Silicene Superstructure

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Silicene is a monolayer of silicon atoms forming a two-dimensional honeycomb lattice. We investigate the topological properties of a silicene superstructure generated by an external periodic potential. The superstructure is a quantum spin-Hall (QSH) insulator if it is topologically connected to silicene. It is remarkable that two inequivalent K and K’ points in the silicene Brillouin zone are identified in certain superstructures. In such a case two Dirac cones coexist at the same Dirac point in the momentum space and they are hexagonally warped by the Coulomb interaction. We carry out a numerical analysis by taking an instance of the (3×3) superstructure on the (4×4) structure of the Ag substrate. We show that it is a QSH insulator, that there exists no topological phase transition by external electric field, and that the hexagonally warping occurs in the band structure.

INTRODUCTION

Topological insulator is a new state of matter which is characterized by an insulating bulk and surface edge modes[1,2]. It is robust against perturbations such as disorders and impurities as far as the gap does not close. It is an interesting and important question whether the topological insulator is robust against a formation of a superstructure in a periodic potential. This is nontrivial since it changes a global structure of the material such as the Brillouin zone. It is intriguing that superstructures have been realized in silicene on top of the Ag substrate[3–5]. There may well be other ways to create periodic potentials. Silicene is an interesting play ground where many types of superstructures are naturally materialized.

Silicene is a sheet of silicon atoms replacing carbons in graphene. It could follow the trend of graphene and attract much attention[3–14]. It would open new perspectives for applications, especially due to its compatibility with Si-based electronics. Almost every striking property of graphene could be transferred to this innovative material. Indeed, it has Dirac cones akin to graphene. It has additionally a salient feature, that is a relatively large spin-orbit (SO) gap, which provides a mass to Dirac electrons and realizes a detectable quantum spin Hall (QSH) effect[8,10]. The QSH insulator is a two-dimensional topological insulator with helical gapless edge modes[1,2]. Furthermore a topological phase transition occurs from the QSH insulator to the trivial band insulator in the electric field[10]. In this paper we address the problem if a superstructure is a QSH insulator as well and if there exists a similar topological phase transition.

In silicene the states near the Fermi energy are π orbitals residing near the inequivalent K and K’ points at opposite corners of the hexagonal Brillouin zone. It is folded into a reduced Brillouin zone in a superstructure. We can generally argue that a superstructure is a QSH insulator provided it is topologically connected to silicene, namely, the gap is open and does not close as the periodic potential is continuously switched off. We also study the problem whether a silicene superstructure undergoes a topological phase transition by applying external electric field as in free-standing silicene[10]. Furthermore, we point out an intriguing possibility that the K and K’ points are identified by the folding. When it happens, there exists only one Dirac point where two Dirac cones coexist, and this is experimentally detectable. In addition to general arguments, to demonstrate these novel phenomena we carry out a numerical analysis by taking an instance of the (3×3) superstructure on the (4×4) structure of the Ag substrate[15].

This paper is composed as follows. In Section II, we present how a superstructure is constructed from a honeycomb lattice. We derive the condition that the K and K’ points coincide. The (3×3) superstructure is the simplest example where this coincidence occurs. In Section III, we postulate the tight-binding Hamiltonian to describe a superstructure. Making a numerical analysis of the band structure of the (3×3) superstructure, we explicitly see that two degenerate Dirac cones are merged into two degenerate hexagonally warped cones in the presence of infinitesimal Coulomb interactions. The mechanism of the hexagonal warping is explained based on the effective low-energy Dirac theory. In Section IV we investigate the topological properties of superstructures in three different ways, i.e., the topological connectedness, the bulk-edge correspondence and the Dirac theory. We present a general criterion when the topological phase transition may occur in superstructures. In the instance of the (3×3) superstructure, it is shown to be topologically connected to silicene, and hence it is a QSH insulator. We also show that there exists no topological phase transition in external electric field. Section V is devoted to discussions.

SUPERSTRUCTURE

We start with a study of a superstructure made on the honeycomb lattice (Fig.1). The honeycomb lattice is specified by the two basis vectors a1 and a2. It has two inequivalent sites (A and B sites) per unit cell. There are three B sites adjacent to one A site. The A and B sites are generated by

\[ A(n_1, n_2) = n_1 a_1 + n_2 a_2, \]

\[ B(n_1, n_2) = n_1 a_1 + n_2 a_2 + r_1, \]

where \( n_1, n_2 \) are integers.
where \( r_i \) is a vector connecting adjacent A and B sites, and \( n_i \) are integers. They span the the A-sublattice and the B-sublattice. A superstructure is specified by two translational vectors \( g_1 \) and \( g_2 \) defined by
\[
g_1 = p \alpha_1 + q \alpha_2, \quad g_2 = e^{\pm i \pi/3} g_1, \tag{2}
\]
with \( p \) and \( q \) integers subject to \( 0 < p \). The choice of the angles \( \pm \pi/3 \) generates the same superstructure due to the hexagonal symmetry. There are two types: The type-I superstructure is generated by choosing \( 0 \leq q \leq p \), while the type-II superstructure is generated by the space inversion of the type-I. When \( q = 0 \), type-I and type-II are identical. We show two examples by choosing \( (p, q) = (3, 0) \) and \( (2, 1) \) in Fig.1.

By a geometrical analysis of the honeycomb lattice we have
\[
|g_1| = \sqrt{p^2 + q^2 + pq}. \tag{3}
\]
The structure thus generated is customarily referred to as the \((|g_1| \times |g_1|)\) superstructure. The number of silicon atoms per unit cell is given by
\[
N = 2 |g_1|^2 = 2 (p^2 + q^2 + pq). \tag{4}
\]
The angle \( \phi \) between the vector \( g_1 \) and the \( y \)-axis is given by
\[
\tan \phi = \sqrt{3} q / (2p + q). \tag{5}
\]
For instance, the choices \( (p, q) = (3, 0) \) and \( (2, 1) \) generate the \((3 \times 3)\) superstructure with \( N = 18 \) and \( \phi = 0 \) and the \((\sqrt{7} \times \sqrt{7})\) superstructure with \( N = 14 \) and \( \phi = \arctan(\sqrt{3}/5) \), respectively, as illustrated in Fig.1.

The basis vectors \( b_i \) of the reciprocal lattice are given by solving the relations \( b_i \cdot \alpha_j = 2 \pi \delta_{ij} \) in the honeycomb lattice (Fig.2). The Brillouin zone is made by \( b_1 \) and \( b_2 \) in the reciprocal lattice with opposite sides being identified. A similar construction is carried out for a superstructure. The reciprocal vectors \( G_1 \) and \( G_2 \) of a superstructure is given by
\[
G_i = (g_i \times n) / |g_1 \times g_2|, \tag{6}
\]
where \( n \) is the unit vector perpendicular to the silicene plane. The Brillouin zone of a superstructure is constructed from that of silicene by identifying two momentum vectors \( k \) and \( k' \) provided they are different only by the principal reciprocal vectors \( G_1 \) and \( G_2 \), as illustrated in Fig.2.

There may occur an intriguing phenomenon. In general, when there are two integers \( n_i \) such that
\[
K - K' = n_1 G_1 + n_2 G_2, \tag{7}
\]
the \( K \) and \( K' \) points are identified. We call it the \( \bar{K} \) point, and choose \( \bar{K} = (0, 0) \). Two Dirac cones coexist at the \( \bar{K} \) point. This happens indeed in the \((3 \times 3)\) superstructure, where the Brillouin zone is obtained by folding that of silicene three times along the \( k_x \)-axis and three times along the \( k_y \)-axis. The area of the Brillouin zone is 1/9 of that of silicene, and its shape is rhombus (Fig.2). This superstructure is simplest and most interesting, which we investigate as an explicit example.

**TIGHT-BINDING HAMILTONIAN**

The superstructure system is described by the second-nearest-neighbor tight-binding model constructed as follows. The Hamiltonian consists of two parts. The basic part is the Hamiltonian of silicene.[16]

\[
H_0 = -t \sum_{\langle i,j \rangle} c_{i \alpha}^\dagger c_{j \alpha} + \frac{\lambda_{SO}}{3 \sqrt{3}} \sum_{\langle \langle i,j \rangle \rangle_{\alpha \beta}} n_{ij} c_{i \alpha}^\dagger \sigma_{\alpha \beta} c_{j \beta}, \tag{8}
\]
where $c^\dagger_{i\alpha}$ creates an electron with spin polarization $\alpha$ at site $i$, and $(i,j)$ and $(i,j')$ run over all the nearest/next-nearest neighbor hopping sites. The first term represents the usual nearest-neighbor hopping with the transfer energy $t = 1.6\text{eV}$. The second term represents the intrinsic SO coupling with $\lambda_{SO} = 3.9\text{meV}$, where $\sigma = (\sigma_x, \sigma_y, \sigma_z)$ is the Pauli matrix of spin, $\nu_{ij} = (d_i \times d_j) / |d_i \times d_j|$ with $d_i$ and $d_j$ the two bonds connecting the next-nearest neighbors. Additionally there exist two types of Rashba SO couplings ($\lambda_{R1}$ and $\lambda_{R2}$) to describe silicene $^{11}$. They are quite small, and play no important roles in the present analysis, as far as we have numerically checked. To avoid unnecessary complications, we neglect them.

The second part is the periodic potential term $H_1$ that introduces a superstructure to the honeycomb lattice. The periodicity is such that the band structure satisfies

$$\varepsilon(k + G_i) = \varepsilon(k), \quad (9)$$

where $G_i$ are the principal reciprocal vectors $^6$. It is to be emphasized that the band structure is simply constructed by folding that of silicene when $H_1 \approx 0$, as illustrated in Fig.3 for an instance of the $(3 \times 3)$ superstructure.

### An Explicit Example and Numerical Results

Superstructures have been materialized on the Ag substrate. The structure of the substrate acts as a periodic potential to silicene and generate a superstructure on silicene. Such an effect can be formulated into the periodic potential term $H_1$.

Silicene available now is mostly grown on the Ag$^{(111)}$ substrate. The most common one is the $(3 \times 3)$ superstructure synthesized on the $(4 \times 4)$ Ag-structure $^3$ $^4$, though many other superstructures are possible due to the commensurability with the Ag substrate $^{17}$ $^{18}$. There are $3 \times 3$ silicon atoms on top of $4 \times 4$ silver atoms in this structure. The unit cell contains $2 \times (3 \times 3) = 18$ silicon atoms. The superstructure contains two sublattices: There are 6 (12) atoms in the higher (lower) sublattice which is $3.0\text{Å} (2.2\text{Å})$ above the Ag surface, as illustrated in Fig.4. This structure has been observed in scanning tunneling microscopy (STM) images, where only the atoms in the higher sublattice are visible $^{11}$ $^{12}$. We may summarize the effect from the Ag substrate as a chemical potential difference $U$ between silicon atoms in higher and lower sublattices. Additionally we may apply the electric field $E_z$ perpendicular to the plane. It generates a staggered sublattice potential $\propto 2\ell E_z$ with $2\ell \approx 0.8\text{Å}$ between them.

The basic nature of the above superstructure is summarized into the periodic potential term,

$$H_1 = U' \sum_{i \in \text{high}} c_{i\alpha}^\dagger c_i, \quad (10)$$

with

$$U' = U + 2\ell E_z, \quad (11)$$

where the summation is taken over higher sites: $U$ is a constant of the order of $0.1\text{eV}$ according to the first-principle calculation $^{18}$. The total Hamiltonian is $H = H_0 + H_1$, where the spin $s_z$ is a good quantum number. Hence, the Hamiltonian is decomposed into the spin sectors indexed by $s_z = \pm 1$. Since the number of silicon atoms per unit cell is 18 in the $(3 \times 3)$ superstructure, we analyze the 18-band tight-binding model. We show numerically calculated band structures of the superstructure for $U' = 0$ and $U' \neq 0$ in Fig.5. They have qualitatively...
Dirac Theory

To understand this phenomenon of the hexagonal warping analytically, we analyze the low energy Dirac theory of the Hamiltonian (3).

\[
H_q = \hbar v_F (k_x \tau_x + \eta k_y \tau_y) + \frac{\eta \alpha e^{3\pi i/2}}{4\sqrt{3}} \left( k_y^2 \sigma_+ + k_x^2 \sigma_- \right) - \eta \lambda_{SO} \sigma_z \tau_z, \tag{12}
\]

where \( \tau = (\tau_x, \tau_y, \tau_z) \) is the Pauli matrix of pseudospin associated with the A and B sites. Two Dirac cones are labelled by \( \eta = \pm \), which are originally present at the K point \((\eta = +)\) and the K’ point \((\eta = -)\) in silicene. The way of trigonal warping is represented by the factor \( e^{3\pi i\eta/2} \) with the phase being opposite \((\eta = \pm 1)\) between the two types of electrons [19]. We assume \( U' = 0 \) in the analytical treatment for simplicity. Nevertheless, we are able to see clearly how two trigonally warped Dirac cones are transformed into two degenerate hexagonally warped Dirac cones.

When we introduce the on-site Coulomb interaction, the Hamiltonian \( H_K \) and \( H_{K'} \) are mixed into

\[
H = \begin{pmatrix}
H_K & H_{12} \\
H_{21} & H_{K'}
\end{pmatrix}, \tag{13}
\]

where we have chosen the basis \( \{ c^K_A, c^K_B, c^{K'}_A, c^{K'}_B \} \), and

\[
H_K = \begin{pmatrix}
-\lambda_{SO} & \hbar v_F k_- + ae^{-3\pi i/2} k_x^2 + \lambda_{SO} \\
\hbar v_F k_+ + ae^{3\pi i/2} k_x^2 & -\lambda_{SO}
\end{pmatrix}, \tag{14}
\]

\[
H_{K'} = \begin{pmatrix}
\lambda_{SO} & \hbar v_F k_- - ae^{-3\pi i/2} k_x^2 + \lambda_{SO} \\
\hbar v_F k_+ - ae^{3\pi i/2} k_x^2 & -\lambda_{SO}
\end{pmatrix}. \tag{15}
\]
TOPOLOGICAL NUMBERS

The topological quantum numbers\[1, 2\] are the Chern number $C$ and the $\mathbb{Z}_2$ index. If the spin $s_z$ is a good quantum number, the $\mathbb{Z}_2$ index is identical to the spin-Chern number $C_s$ modulo 2. They are defined when the state is gapped and given by

$$C = C_\uparrow + C_\downarrow, \quad C_s = \frac{1}{2}(C_\uparrow - C_\downarrow),$$

where $C_{\uparrow \downarrow}$ is the summation of the Berry curvature in the momentum space over all occupied states of electrons with $s_z = \pm 1$. The characteristic feature is that they are unchanged even if some parameters are continuously switched off in the Hamiltonian provided that the gap does not close\[20, 21\].

The spin-Chern number is defined by the integration of the Berry curvature $\Omega_i(k)$ over the conduction band indexed $i$ in the Brillouin zone $S$ for each spin sector,

$$C_{s_z} = \sum_{i=1}^{N/2} \int_S d^2k \Omega_i(k).$$

Here, the Brillouin zone $S$ is that of the superstructure. The number of conduction bands is $N/2$ when the system is half filled. By making use of the band structure\[9\] we may equivalently integrate the Berry curvature $\Omega(k)$ over one conduction band in the extended Brillouin zone $S_{ex}$, which is the Brillouin zone of silicene,

$$C_{s_z} = \int_{S_{ex}} d^2k \Omega(k).$$

Recall that $N/2$ conduction bands in the Brillouin zone $S$ are merged into one conduction band in the extended Brillouin zone $S$, as illustrated in Fig\[3\].

It is well known that silicene is a SQH insulator characterized by the topological numbers,

$$C = 0, \quad C_s = 1.$$

It is an important question whether a superstructure is also a topological insulator. We investigate this problem in the following three ways. All of them produce the same result on this problem.

Adiabatic Analysis

We take the example of the $(3 \times 3)$ superstructure by investigating our Hamiltonian system $H = H_0 + H_1$. We have explicitly calculated the band gap of the $(3 \times 3)$ superstructure by taking the potential term (10), whose result is given in Fig\[8\]. It is surprising that the band gap never closes as a function of $U' = U + 2\epsilon c_r$, Fig. 8. It is well known that silicene is a SQH insulator characterized by the topological numbers,

$$C = 0, \quad C_s = 1.$$

When we apply external electric field $E_z$, silicene undergoes a topological phase transition\[10\]. However, the $(3 \times 3)$ superstructure does not since the gap does not close. We shall see the reason why this is the case in subsection Edges.

Edge States Analysis

The bulk-edge correspondence\[1, 2\] is well known to characterize the topological insulators. Namely, a topological insulator is characterized by the appearance of gapless modes on edges. The reason why gapless modes appear in the edge of a topological insulator is understood as follows. The topological insulator has a nontrivial topological number, the $\mathbb{Z}_2$ index\[16\], which is defined only for a gapped state. When a topological insulator has an edge beyond which the region has the trivial $\mathbb{Z}_2$ index, the band must close and yield gapless modes in the interface. Otherwise the $\mathbb{Z}_2$ index cannot change its value across the interface.

We have numerically calculated the band structure of a zigzag nanoribbon geometry in Fig\[3\](a,b), where the width
The width contains 8 unit cells of the superstructure. Two zero-energy edge modes (indicated by red lines) are clearly seen: Each line represents the two-fold degenerate helical modes propagating one edge as indicated in Fig. 9(c). The band structure of a nanoribbon with \( U' = 0 \) is just the one obtained by folding that of a silicene nanoribbon as in Fig. 9(a). It is modified for \( U' \neq 0 \), but the modification is only slight even when we take \( U' = 0.5t \). We set \( t_{\perp \parallel} = 1.5t \) and \( t_{\parallel \parallel} = t \). By comparing these with Fig. 6, the modification of the band structure is found to be quite small.

![Figure 9: Band structure of a zigzag nanoribbon made of the (3 x 3) superstructure (a) for \( U' = 0 \) and (b) for \( U' = 0.5t \). The width contains 8 unit cells of the superstructure. The red lines indicate the doubly degenerate edge modes with up and down spins. The vertical axis is the energy in unit of \( t \). The horizontal axis is the momentum \( k \). The points \( k = 0 \) and \( k = \pi \) are identified, and give the \( \bar{k} \) point, while \( k = \pi \) is the \( \Gamma \) point. We have set \( \lambda_{SO} = 0.2t \) for illustration. Each red line connects the \( \bar{k} \) point in the conduction and valence bands after winding once a circle of the whole Brillouin zone. (c) Illustration of a nanoribbon with the width containing 3 unit cells of the superstructure. The red and blue arrows indicate the degenerate gapless edge modes with up and down spins.](image)

**DISCUSSIONS**

In conclusion, we have investigated the topological properties of a superstructure made from the silicene honeycomb.
The superstructure is a QSH insulator if it is topologically connected to silicene. The concept of the topological connectedness may be formulated as follows. We consider the Hamiltonian,

$$H(\lambda) = H_0 + \lambda H_1.$$  \hspace{1cm} (25)

Provided the gap is open in the system $H(\lambda)$ for any values of $\lambda$ ($0 \leq \lambda \leq 1$) as $\lambda$ is continuously changed from $\lambda = 1$ to $\lambda = 0$, the two systems $H(1)$ and $H(0)$ are topologically connected. For the instance of the potential (10) we have used the electric field $E_z$ instead of $\lambda$ as such a continuous parameter to show the topological connectedness.

We have investigated the problem if the topological phase transition occurs as in silicene. We have also derived the condition (7) that the K and K’ points become identical in the transition occurs as in silicene. We have carried out a numerical analysis and confirm these observations by taking an instance of the paper [10].

Silicene superstructures are grown on the Ag substrate. The most common one is the $(3 \times 3)$ superstructure on the $(4 \times 4)$ Ag-structure. There are several possible superstructures obtained in this way. Here we give some correspondences between them:

| Si-superstructure | Ag-structure | $p/q$ ratio | Refs. |
|-------------------|--------------|-------------|-------|
| $3 \times 3$      | $4 \times 4$ | 3/0 1.33    | [3,4] |
| $2 \times 2$      | $\sqrt{2} \times \sqrt{2}$ | 2/0 1.32   |       |
| $\sqrt{7} \times \sqrt{7}$ | $13 \times 13$ | 2/1 1.36 | [4]   |
| $2\sqrt{3} \times 2\sqrt{3}$ | $2\sqrt{21} \times 2\sqrt{21}$ | 2/2 1.32 |       |
| $\sqrt{7} \times \sqrt{7}$ | $2\sqrt{3} \times 2\sqrt{3}$ | 2/1 1.31 | [17]  |

It is interesting that the ratios of the Ag-structure and Si-superstructures are around 1.3. There would be other ways to construct superstructures by introducing periodic potentials to silicene. Our analysis of topological properties in superstructure is applicable to any of them.

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