Formation of quantum spin Hall state on Si surface and energy gap scaling with strength of spin orbit coupling

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For potential applications in spintronics and quantum computing, it is desirable to place a quantum spin Hall insulator [i.e., a 2D topological insulator (TI)] on a substrate while maintaining a large energy gap. Here, we demonstrate a unique approach to create the large-gap 2D TI state on a semiconductor surface, based on first-principles calculations and effective Hamiltonian analysis. We show that when heavy elements with strong spin orbit coupling (SOC) such as Bi and Pb atoms are deposited on a patterned H-Si(111) surface into a hexagonal lattice, they exhibit a 2D TI state with a large energy gap of $0.5 \text{ eV}$. The TI state arises from an intriguing substrate orbital filtering effect that selects a suitable orbital composition around the Fermi level, so that the system can be matched onto a four-band effective model Hamiltonian. Furthermore, it is found that within this model, the SOC gap does not increase monotonically with the increasing strength of SOC. These interesting results may shed new light in future design and fabrication of large-gap topological quantum states.

Recently there has been a surge in the investigation of topological insulators (TIs). TIs are characterized by topologically protected metallic surface or edge states with helical spin polarization residing inside an insulating bulk gap. These states have negligible elastic scattering and Anderson localization, which may provide ideal dissipationless spin current for future electronic devices with low power consumption. To realize their potential applications, it is desirable for the TIs to have an energy gap as large as possible, i.e., for room temperature applications. As for 2D TIs [i.e., quantum spin Hall (QSH) insulators], they also need to be grown or placed onto a substrate or formed as an interface, while maintaining a large gap. One desired approach is to directly fabricate large-gap TIs on semiconductor surfaces, which may avoid problems like transfer or interfacing a 2D layer over a foreign substrate. So far, however, this goal remains allusive.

The HgTe quantum well, as the first theoretically predicted and experimentally confirmed QSH insulator, has a small gap of 40 meV with topological edge states only detectable at very low temperature ($<10 \text{ K}$). Recent studies pertaining to Bi/Sb(111) films, Sn films, metal-decorated graphene, silicon/germanene, and 2D organometallic frameworks have largely enriched the family of 2D TIs, and some of them have a large gap. However, a critical drawback with most previous theoretical studies of 2D TIs is their reliance on the electronic and topological properties of freestanding films, whose existence can be in doubt. Even a freestanding film does exist, its properties are expected to be influenced by the underlying substrate in real applications.

Here, we demonstrate a unique approach of creating QSH state on a conventional semiconductor surface via depositing heavy elements with strong spin orbit coupling (SOC) onto a patterned Si(111) surface into a hexagonal lattice, which exhibit TI state with a large energy gap of $\sim 0.5 \text{ eV}$. Here, the substrate plays a ‘positive’ role acting as an orbital filter to critically select the orbital composition around the Fermi level to realize nontrivial large-gap topological state. Specifically, the surface system can be matched onto an effective four-band model Hamiltonian which captures the underlying physics. We depict a unified picture of energy gap as a function of SOC to achieve large-gap QSH state. Importantly, we found that it is not necessarily true to have a large gap with a heavier atom of larger SOC, a noteworthy point for future design of TIs.

We have performed density functional theory (DFT) based first-principles calculations of band structure and band topology of 2D hexagonal lattices of various metal atoms, including Bi, Pb, Sb, Sn, Ga, In and TI, grown on a patterned H-saturated Si(111) surface. The detailed methodologies are presented in the Supplementary Information. We will first discuss in detail the results of Bi and Pb, as representative examples, and leave the
results of other metals for later discussion. Atomically flat H-Si(111) surface has been prepared for decades and is a widely-used substrate for epitaxial growth of ordered overlayers. The surface dangling bonds are passivated by H to avoid surface reconstruction. In order to form a hexagonal metal overlayer lattice, we propose a two-step fabrication process, as shown in Fig. 1. First, to create a desirable surface template pattern for metal growth, H atoms are selectively removed in hexagonal symmetry using scanning tunneling microscopy, as discussed in Refs. 30, 31; Second, heavy metal atoms with large SOC can be deposited to grow or self-assemble into the exposed Si sites, as already demonstrated for other systems.

We found a very strong binding between the deposited metal atoms and the exposed Si atoms in the H-Si(111) surface, as evidenced by the calculated adsorption length \(d\) (see Fig. 1(c)) of 2.68 Å and 2.75 Å for Bi and Pb, respectively. The high structural stability is also indicated by a large adsorption energy \(E_{\text{ad}}\) (see Fig. 1(c)) of 2.68 Å and 2.75 Å for Bi and Pb, respectively. The upper Dirac band largely overlaps with the conduction band, as in H-Si(111). Analysis of band composition further is also indicated by a large adsorption energy (see Fig. 1(c)) of 2.68 Å and 2.75 Å for Bi and Pb, respectively. The adsorption energy is found to be 3.88 eV and 3.92 eV for Bi and Pb, respectively, which are much larger than the binding energies of bulk Bi (2.18 eV) and Pb (2.03 eV) in the crystalline solid. The top and side view of the proposed structure, with the surface unit cell vector \(a_1\) and \(a_2\) indicated in (b) and the adsorption length \(d\) in (c). (d) The first surface Brillouin zone.

Figure 1 | A two-step approach to fabricate 2D TI by deposition of heavy metal atoms on a patterned H-Si(111) surface. (a) Schematic view. (b, c) The top and side view of the proposed structure, with the surface unit cell vector \(a_1\) and \(a_2\) indicated in (b) and the adsorption length \(d\) in (c). (d) The first surface Brillouin zone.

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SCIENTIFIC REPORTS | 4 : 7102 | DOI: 10.1038/srep07102
Figure 2 | Band structures of Bi and Pb@H-Si(111). (a–b) Without SOC. The Fermi level is set at zero. The green (yellow) shaded area represents the valence bands (conduction bands) of Si. Band compositions around Fermi level are also indicated. (c–d) Same as (a–b) with SOC.

Figure 3 | Electronic structures of Bi@H-Si(111) and its edge state. (a) Comparison of band structures for Bi@H-Si(111) calculated by DFT (black lines) and Wannier function method (green circles). (b) The Dirac edge states within the SOC-induced band gap. Scale bar is indicated on the right. (c) The partial DOS projected onto \( p_x \), \( p_y \), and \( p_z \) orbitals of Bi, and the total DOS of neighboring Si atoms. (d) Top: The charge density redistribution induced by metal atom surface adsorption for Bi@H-Si(111) (isovalue = 0.02 e/Å\(^2\)), illustrating saturation of Bi \( p_z \) orbital. Bottom: Same as Top for the H-saturated freestanding planar Bi lattice.
$Z_2 = 0$ (see Fig. S3). When it is placed onto the H-Si(111) surface or adsorbed with H, it becomes topologically nontrivial (Figs. S1 and S4). This originates from the intriguing orbital filtering effect imposed by the substrate or H saturation, which selectively remove the $p_z$ orbitals from the Bi lattice to realize the large-gap QSH phase.

Specifically, we can describe the Bi@H-Si(111) using a simplified ($p_x$, $p_y$) four-band model Hamiltonian in a hexagonal lattice as $^{24,38}$,

$$H_{\text{eff}} = \begin{pmatrix}
    \epsilon_0 & 0 & S_{xx} & S_{xy} \\
    0 & \epsilon_0 & S_{xy} & S_{yy} \\
    S_{xx} & S_{xy} & \epsilon_0 & 0 \\
    S_{yy} & S_{xy} & 0 & \epsilon_0
\end{pmatrix} + \sigma_2 \lambda_{so} \begin{pmatrix}
    0 & -i & 0 & 0 \\
    i & 0 & 0 & 0 \\
    0 & 0 & 0 & -i \\
    0 & 0 & i & 0
\end{pmatrix},$$

in which $S_{xx} = V_{ppx} + \left(1/4 \right) V_{ppy} + \left(3/4 \right) V_{ppz}$, $S_{xy} = 3/4 \left(\epsilon_{1a}^x + \epsilon_{1a}^y \right)$, $S_{yy} = V_{ppx} + \left(3/4 \right) V_{ppy} + \left(1/4 \right) V_{ppz}$, $\epsilon_{1a}^x$, $\epsilon_{1a}^y$, $\epsilon_{1a}^z$, $\epsilon_{2a}^x$, $\epsilon_{2a}^y$, $\epsilon_{2a}^z$, $\epsilon_{3a}^x$, $\epsilon_{3a}^y$, $\epsilon_{3a}^z$ are the lattice vectors, $V_{ppx}$, $V_{ppy}$, and $V_{ppz}$ are the Slater-Koster parameter $^{39}$, and $\sigma_2 = \pm 1$ is the spin eigenvalue.

Diagonalization of Eq. (1) in reciprocal space gives the band structures shown in Fig. 4, which shows typical four bands as a function of SOC strength ($\lambda$) scaled by $t$ ($t$ is the coupling strength between neighboring $p_x$ and $p_y$ orbitals). Fermi energy is set to zero. The SOC induced energy gaps ($\Delta E_1$, $\Delta E_2$, and $\Delta E_3$) are indicated. The global gap transition from $K$ point to $\Gamma$ point driven by SOC can be clearly seen.

Figure 4 | Energy bands resulting from the four-band model [Eq. (1)] as a function of SOC strength ($\lambda$) scaled by $t$ ($t$ is the coupling strength between neighboring $p_x$ and $p_y$ orbitals). Fermi energy is set to zero. The SOC induced energy gaps ($\Delta E_1$, $\Delta E_2$, and $\Delta E_3$) are indicated. The global gap transition from $K$ point to $\Gamma$ point driven by SOC can be clearly seen.
deposition of different metal atoms on different substrates.\textsuperscript{11,27} It opens up a new and exciting avenue for future design and fabrication of room temperature topological surface/interface states based on current available epitaxial growth and semiconductor technology, where the metal overlayer is atomically bonded but electronically isolated from the underneath semiconductor substrate.\textsuperscript{27}

**Methods**

Our electronic structure calculations based on density functional theory were performed by using a plane wave basis set\textsuperscript{41} and the projector-augmented wave method\textsuperscript{42}, as implemented in the VASP code\textsuperscript{43}. The exchange-correlation functional was treated with the generalized gradient approximation in Perdew-Burke-Ernzerhof format\textsuperscript{44}. Calculations of $Z_2$ triviality were carried out by using the full-potential linearized augmented plane-wave method implemented in the WIEN2K package\textsuperscript{45}. Details for models and computations ($Z_2$ invariant calculation results, and band structures of Sb and Sn@H-Si(111)) are presented in Supplementary Information.

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**Figure 5** Band structures of Ga, In, Tl@H-Si(111). (a–b) Band structures of Ga@H-Si(111) without and with SOC, respectively. The Fermi level is set to zero. Band compositions around Fermi level are indicated. (c–d) Same as (a–b) for In@H-Si(111). (e–f) Same as (a–b) for Tl@H-Si(111).
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