Experimental demonstration of the topological surface states protected by the time-reversal symmetry

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The strong spin-orbital coupling in a certain class of materials gives rise to the novel topological insulators in two 1, 2 and three dimensions 3, 4, 5, 6, 7 in the absence of an external magnetic field. The topological states on the surfaces of three dimensional (3D) materials have been studied recently in Bi1−xSbx 8, 9, 10, Bi2Te3 and Bi2Se3 11, 12, 13, which possess insulating gaps in the bulk and gapless states on surfaces. The surface states of a 3D topological insulator comprise of an odd number of massless Dirac cones and the crossing of two dispersion branches with opposite spins is fully protected by the time-reversal-symmetry at the Dirac points. Such spin-helical states are expected to bring forward exotic physics, such as magnetic monopole 14 and Majorana fermions 15, 16. To date, the experimental study of topological insulators is predominantly limited to the determination of their band structure by angle-resolve photoemission spectroscopy (ARPES) 8, 9, 10, 11, 12, 13. Distinct quantum phenomena associated with the nontrivial topological electronic states still remain unexplored. Particularly, there is no direct experimental evidence for the time reversal symmetry that protects the topological property. Here, using the low temperature scanning tunneling microscopy (STM) and spectroscopy (STS), we report the direct observation of quantum interference caused by scattering of the 2D topologically nontrivial surface states off impurities and surface steps. Our work strongly supports the surface nature of the topological states, and provides a way to study the spinor wave function of the topological state. More significantly, we find that the backscattering of topological states by a nonmagnetic impurity is forbidden. This result directly demonstrates that the surface states are indeed quantum-mechanically protected by the time reversal symmetry.

The interference patterns in STM experiments 17, 18 result from the 2D surface states perturbed by surface defects. A surface state is uniquely characterized by a 2D Bloch wave vector \( \vec{k} \) within the surface Brillouin zone (SBZ). During elastic scattering, a defect scatters the incident wave with a wave vector \( \vec{k}_i \) into \( \vec{k}_f = \vec{k}_i + \vec{q} \), with \( \vec{k}_i \) and \( \vec{k}_f \) being on the same constant-energy contour (CEC). The quantum interference between the initial and final states results in a standing wave pattern whose spatial period is given by \( 2\pi/\vec{q} \). When the STM images of a standing wave are Fourier transformed 21, the scattering wave vector \( \vec{q} \) is the momentum transfer) becomes directly visible in the reciprocal space. In contrast, for bulk states, there will be continuous ranges of wave vectors on the projected SBZ for a given energy. Usually, no distinct interference fringe can be produced by bulk states and visualized by STM. In this sense, the standing wave is surface-states-sensitive and particularly suitable for studying topological insulators.

Our experiments were conducted in an ultra-high vacuum low temperature (down to 0.4 K) STM system equipped with molecular beam epitaxy (MBE) for film growth (Unisoku). The stoichiometric Bi2Te3 film, a robust topological insulator, was prepared on single crystal substrate Si(111) by MBE. Details of sample preparation are described elsewhere 22. Shown in Fig. 1(a) is a typical STM image of the Bi2Te3 film with a thickness of \( \sim100 \) nm. The atomically flat morphology of the film is clearly observed. The three steps seen in Fig. 1(a) all have the height (0.94 nm) of a quintuple layer. The steps are preferentially oriented along the three close-packing ([100], [110] and [010]) directions. The image with atomic resolution [Fig. 1(b)] exhibits the two-dimensional hexagonal lattice structure of the Te-terminated (111) surface of Bi2Te3. Our STM observation further reveals a small density of clover-shaped defects on the surface (see supporting material 23). Simi-
the center (\( \bar{\Gamma} \) point) of the SBZ \([10, 11]\), giving rise to a vanishing DOS in the vicinity of topological states of Bi\(_2\)I. According to the calculations \( \text{Fig. 1(d)} \), the contribution of the bulk insulating gap linearly depends on the bias and is attributed to the gapless surface states. These features in STS are in good agreement with those obtained by the first-principles calculations \( \text{see supporting material [23]} \). The atomic resolution image \( \text{Figures 2(c) to 2(g)} \) summarize the \( dI/dV \) maps for bias voltages ranging from 50 mV to 400 mV from the area shown in \( \text{Fig. 2(a)} \). The first striking aspect of these images is the existence of standing wave \([17, 18, 19]\) in the vicinity of the Ag trimmers. The spatial modulation of LDOS by an Ag atom forms a hexagonal pattern, whose edges are perpendicular to the \( \bar{\Gamma} – \bar{M} \) directions in SBZ. This situation is more clearly resolved at large bias voltages \( \text{Figs. 2(f) and 2(g)} \). As expected, the interference pattern is anisotropic as a result of the hexagram CEC \([10]\). The spatial period of the standing wave scales inversely with the bias voltage and is determined by the momentum transfer during scattering at a given energy. Below 50 meV, the fringes become obscured. It results from a combination of two effects: (i) The wavelength increases rapidly as the bias voltage approaches the Dirac point, where \( k = 0 \). (ii) At low energy, more topological surface states with different wavelengths are involved in the formation of standing wave as indicated by the first-principles calculations \( \text{Fig. 1(d)} \). The superposition of waves with various wavelengths smear out the interference fringes. With increasing bias, especially after the surface states in the \( \bar{\Gamma} – \bar{M} \) direction merge into the bulk conduction band at \( \sim 0.2 \text{ eV} \) above the Dirac point according to calculation \( \text{Fig. 1(d)} \), the contribution of states in the \( \bar{\Gamma} – \bar{M} \) direction vanishes and the states in the vicinity of \( \bar{\Gamma} – \bar{K} \) direction gradually gain more weight, leading to more distinct interference patterns. After the surface states in the \( \bar{\Gamma} – \bar{K} \) direction merge into the bulk conduction band at \( \sim 0.6 \text{ eV} \) above the Dirac point \( \text{Fig. 1(d)} \), the standing waves fade out again.

To quantify the standing waves and obtain the scattering wave vectors, we performed Fourier transformation of the \( dI/dV \) maps into the \( \vec{q} \)-space \( \text{Figs. 2(h) to 2(l)} \). One important feature in the power spectra can be immediately discerned by comparing the six-fold symmetric pattern in the \( \vec{q} \)-space with SBZ (the red hexagon in \( \text{Fig. 2(h)} \)); the regions with high intensity are always oriented toward the \( \bar{\Gamma} – \bar{M} \) directions, while the

FIG. 1: (a) The STM topograph of the Bi\(_2\)Te\(_3\)(111) film. The imaged area, 250 nm by 250 nm, was scanned at a sample bias of 3 V and tunneling current of 50 pA. (b) The atomic-resolution image. Tellurium atom (pink colored) spacing is about 4.3 Å. The image was scanned at a sample bias of -40 mV and tunneling current of 0.1 nA. The arrows indicate the bottom of the conduction band and the top of the valence band, respectively. (c) \( dI/dV \) spectrum taken on bare Bi\(_2\)Te\(_3\)(111) surface. The spectrum was measured with setpoint \( V=0.3 \text{ V} \), \( I=0.1 \text{ nA} \). The arrows indicate bulk energy gaps. The surface states are red lines around the \( \bar{\Gamma} \) point.

According to the calculations \( \text{Fig. 1(d)} \), the standing waves fade out again. Below 50 meV, the fringes become obscured. It results from a combination of two effects: (i) The wavelength increases rapidly as the bias voltage approaches the Dirac point, where \( k = 0 \). (ii) At low energy, more topological surface states with different wavelengths are involved in the formation of standing wave as indicated by the first-principles calculations \( \text{Fig. 1(d)} \). The superposition of waves with various wavelengths smear out the interference fringes. With increasing bias, especially after the surface states in the \( \bar{\Gamma} – \bar{M} \) direction merge into the bulk conduction band at \( \sim 0.2 \text{ eV} \) above the Dirac point according to calculation \( \text{Fig. 1(d)} \), the contribution of states in the \( \bar{\Gamma} – \bar{M} \) direction vanishes and the states in the vicinity of \( \bar{\Gamma} – \bar{K} \) direction gradually gain more weight, leading to more distinct interference patterns. After the surface states in the \( \bar{\Gamma} – \bar{K} \) direction merge into the bulk conduction band at \( \sim 0.6 \text{ eV} \) above the Dirac point \( \text{Fig. 1(d)} \), the standing waves fade out again.
in the reciprocal space [Fig. 3(a)]. Generally, the intensity in the $\bar{\Gamma} - \bar{K}$ directions vanishes (see supporting material [22]). Such phenomena can be understood by exploring possible scattering processes on the CEC in the reciprocal space [Fig. 3(a)]. Generally, the $\vec{k}_i$ and $\vec{k}_f$ pairs with high joint DOS should dominate the quantum interference. For energies at which the interference fringes are prominent, the regions on CEC with high DOS are primarily centered around the $\bar{\Gamma} - \bar{K}$ directions [10]. Therefore, three scattering wave vectors, labeled $\vec{q}_1$, $\vec{q}_2$ and $\vec{q}_3$, might be expected to appear in the power spectra. Among them, however, only $\vec{q}_2$ is along the $\bar{\Gamma} - \bar{M}$ directions and can generate the observed standing waves. Both $\vec{q}_1$ and $\vec{q}_3$ are invisible in the power spectra. There is a simple argument to account for the disappearance of $\vec{q}_1$: the time-reversal invariance. The topological states $|\vec{k}_i \rangle$ and $| -\vec{k}_i \rangle$ are related by the time-reversal transformation: $| -\vec{k}_i \rangle = T|\vec{k}_i \rangle$, where $T$ is the time-reversal operator. It is straightforward to show that $\langle -\vec{k}_i | \vec{U} | \vec{k}_i \rangle = -\langle \vec{k}_i | \vec{U} | -\vec{k}_i \rangle^* = -\langle -\vec{k}_i | \vec{U} | \vec{k}_i \rangle = 0$ for fermions, where $\vec{U}$ is a time-reversal invariant operator and represents the impurity potential of the non-magnetic Ag impurity. Therefore, the backscattering between $\vec{k}$ and $-\vec{k}$ is quantum-mechanically prohibited. Most of the observed features in the interference pattern, including the extinction of wave vector $\vec{q}_3$, have been recently well explained by a full theoretical treatment [27] based on the T-matrix approach for multiband systems [28]. In addition to the existence of standing waves, the absence of backscattering represents the second and most striking aspect of our experiment, which makes the topological standing waves more extraordinary as compared to the conventional surface states on metal samples [17, 18, 13, 20].

We can obtain the dispersion of the massless Dirac fermions in the $\bar{\Gamma} - \bar{K}$ direction using the interference patterns and their Fourier transforms. For $\vec{q}_2$, the scattering geometry determines $q_2 = \sqrt{3}k$ [see Fig. 3(a)], where $k$ is the wave vector in the $\bar{\Gamma} - \bar{K}$ direction at a given energy. The resulting $k$ values vary linearly with energy [Fig. 3(b)]. The slope of the linear fitting provides a measurement of the Dirac fermion velocity $v_F$, which is $4.8 \times 10^5$ m/s. In addition, the energy of the Dirac point is estimated to be $0.25$ eV by the intercept of the dispersion with the energy axis. These observations are in agreement with the results from the first-principles calculation and the ARPES data [7, 22]. More importantly, the unoccupied states, which are inaccessible to ARPES,
can be probed by the standing waves with STM.

Interference fringes are also found at the step edges on the surface (29) [Figs. 4(a) to 4(h)]. Similar to the case of Ag trimmers, the standing waves produced by steps are predominantly propagating along the $\bar{\Gamma} - \bar{M}$ direction. The fringes are clearly visible even at the negative bias voltages probably owing to the stronger scattering potential compared to that of the Ag trimmers. The dispersion curve deduced from these patterns again shows a linear relation between the scattering wave vector and the energy [Fig. 4(i)]. Using the slope of the linear fitting together with the same scattering geometry as that for the Ag trimmer, the Fermi velocity is found to be $4.8 \times 10^5$ m/s, the same as that obtained from the standing waves caused by the Ag impurities.

The existence of standing wave strongly supports the surface nature of topological states. An important issue that immediately arises is whether the topological states respond differently to the magnetic and the nonmagnetic impurities. Theoretically, it was pointed out (30, 31) that a time-reversal breaking perturbation, such as magnetic impurities, can induce scattering between the states $|\vec{k}\uparrow\rangle$ and $-|\vec{k}\downarrow\rangle$ and open up a local energy gap at the Dirac point. It remains an open question to observe the distinct signature of time-reversal breaking in topological insulators.

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Note added. At the completion of this manuscript for submission, we became aware of related work by P. Roussan et al. (32). The authors reported STM study of scattering from disorder in BiSb alloy.

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FIG. 4: Standing waves on the upper terrace by a step edge ((a) to (h)). All the images are $dI/dV$ maps at various bias voltages of an area of 35 nm by 35 nm. Imaging conditions: I=0.1 nA. (i) Energy dispersion deduced from the standing waves at the step edge. The dispersion is a function of the scattering wave vector $q$. The inserted STM image shows the step that produces the standing waves in (a) to (h).

The other candidate model of the Ag trimmer is that the Ag atom substitutes a topmost layer Te atom. In this case, the formation of Ag trimmers is kinetically more difficult. Although the exact structure of Ag trimmers does not affect the main conclusion here, it remains an interesting subject for further study, for example, by first-principles calculation.

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