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Non-degenerate metallic states on Bi(114): a one-dimensional topological metal

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The (114) surface of the semimetal Bi is found to support a quasi one-dimensional, metallic surface state. As required by symmetry, the state is degenerate along the $\bar{\Gamma}-\bar{Y}$ line of the surface Brillouin zone with a highest binding energy of ≈150 meV. In the $\bar{\Gamma}-\bar{X}$ direction the degeneracy is lifted by the strong spin-orbit splitting in Bi, as directly shown by spin-resolved photoemission. This results in a Fermi contour consisting of two closely separated, parallel lines of opposite spin direction. It is argued that similar states on related insulators would give rise to a one-dimensional quantum spin Hall effect.

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Surface and interface states on so-called topological insulators have recently attracted considerable attention because of their role in the quantum spin Hall effect [1–5]. The strong spin-orbit interaction together with the particular topology of the valence and conduction band states involved give rise to a robust gapless excitation spectrum at the surface and on the edges of such crystals. The term 'topological metal' has been coined to describe these states [6, 7]. Such states are a prerequisite for the formation of topological insulators. Examples of topological insulators are bismuth/antimony alloys with certain special concentrations (e.g. $\mathrm{Bi}_{0.9}\mathrm{Sb}_{0.1}$) [8, 9]. For the $\mathrm{Bi}_{0.9}\mathrm{Sb}_{0.1}$(111) surface, angle-resolved photoemission has revealed the insulating character of the bulk as well as the existence of two-dimensional metallic surface states. These states have been shown to be topologically non-trivial, i.e. they cannot be removed by a small perturbation [8].

In the quantum spin Hall effect the 'topologically metallic' edge states play a similar role as the edge states in the ordinary quantum Hall effect. However, their character and symmetry is fundamentally different: each edge state is actually a pair of states which are connected by time-reversal symmetry and have opposite spin and k. Unlike in the ordinary quantum Hall effect, the spin quantum Hall effect does not break time-reversal symmetry and thus exists (only) in the absence of an external magnetic field. Using a combination of experimental techniques, we show in this paper that a vicinal surface of bismuth, Bi(114), supports a one-dimensional and spin-split electronic state. The electronic structure at the Fermi energy consists of only two crossings with opposite spin and k and therefore strongly resembles the edge states in the quantum spin Hall effect.

For the present experiment we use pure Bi rather than a BiSb alloy because this provides the possibility to study a larger variety of surface orientations. It can be expected [6, 7, 9], and it was found by comparing Bi(111) [10] with $\mathrm{Bi}_{0.9}\mathrm{Sb}_{0.1}$(111) [8], that the surface states of the alloy are very similar to those of Bi single crystals [10–13]. We argue that the one-dimensional state reported here can be expected to be topologically stable on a corresponding BiSb insulator.

The Bi(114) crystal was mechanically and electrochemically polished. It was cleaned *in situ* by cycles of noble gas sputtering and annealing between 300 and 400 K. Scanning tunneling microscopy (STM) measurements were performed in a low-temperature STM operating at 5 K. Spin-integrated angle-resolved photoemission measurement were performed at the SGM-3 beamline of the storage ring ASTRID [14] at a temperature of ≈ 60 K with an angular resolution 0.2° and an energy resolution of 65 meV and 25 meV for photon energies of 70 eV and 20 eV, respectively. Spin-resolved photoemission experiments were carried out at the COPHEE set-up at the Swiss Light Source, Paul Scherrer Institut, [15] at room temperature, with a photon energy of 21.2 eV and with an energy and angular resolution of 80 meV and 1.5°. Surface cleanliness and order were checked by STM, Auger electron spectroscopy and photoemission (when available) and by low energy electron diffraction (LEED).

The structure of Bi(114) as studied by STM on different length scales is shown in Fig. 1, together with a comparison to structural models. The only symmetry element of the surface is a mirror plane in the y direction.

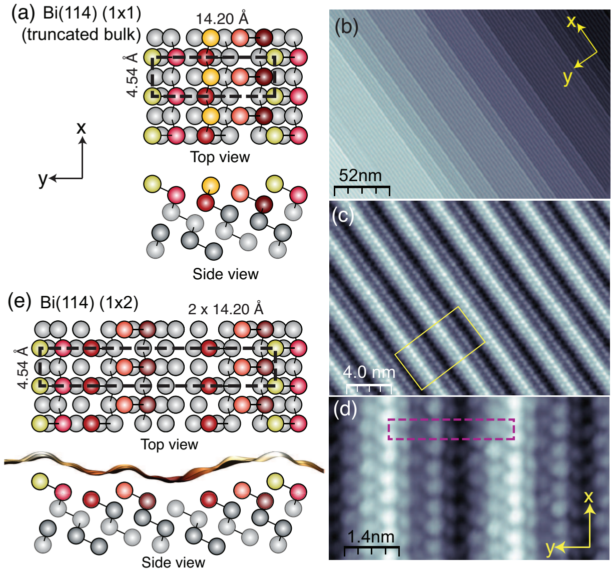

FIG. 1: (Color online) (a) Truncated bulk structure of the Bi(114) surface. The pseudo-covalent bonds of the bulk bismuth double layer structure are indicated as solid lines, for the sake of clarity only some are shown. (b-d) STM images (sample bias 54 mV, -150 mV and -40 mV, respectively). The zoomed image (d) is from the region marked with the yellow rectangle in (c). The unit cell in (d) is also indicated in the atomic model of the (1×2) surface shown in top and side views in (e). Panel (e) also shows the projected STM topography along the x direction.

On a larger scale, the surface appears quite regular with straight atomic rows running perpendicular to the mirror plane, i.e. in the x direction [Figs. 1(b,c)]. Atomically resolved STM images [Figs. 1(c,d)] cannot be reconciled with the truncated bulk structure of Fig. 1(a). Whilst the distance within the atomic rows along the x direction fits with the expected value, the surface shows a larger than expected corrugation along the y direction and the periodicity is twice that of the truncated bulk. Based on high resolution images such as Fig. 1(d), an atomic model of the surface can be constructed simply by removing 4 of the highest lying atomic rows of a doubled (1×2) unit cell [Fig. 1(e)]. The result is a trenched surface with two protruding atomic rows per unit cell. The strong (1×2) reconstruction of Bi(114) significantly enhances the intrinsic one-dimensional character by increasing the periodicity in the y direction to 2.8 nm.

The strongly one-dimensional character of the surface is also reflected in its electronic structure. Figure 2(a) shows the photoemission intensity at the Fermi energy E_F, which can be interpreted as an image of the surface Fermi contour. The dominating feature is a line of high intensity passing through the center of the surface Brillouin zone (SBZ). The corresponding line is also found in the next SBZ. The existence and position of these lines in $\mathbf{k}$ is independent of the photon energy, suggesting the absence of $k_\perp$-dispersion and thus a surface-localized character. In fact, the straight-line shape of the Fermi contour along the y direction is indicative of a purely one-dimensional de-localization in the x direction, consistent with the strongly anisotropic geometrical structure [16, 17]. In addition to the Fermi contour lines, very weak and broad features are found which cut diagonally through the Fermi contour. They do not follow the periodicity of the SBZ and are therefore interpreted as bulk-related.

Figure 2(b) shows an energy vs. k cut along the red line in the Fermi contour image. The states that give rise to the two parallel lines in Fig. 2(a) are clearly identified as small features at E_F, on top of Λ-shaped pairs of bands. The slope of the Λ-shaped bands differs for $k_x \approx 0$ Å^{-1} and ≈1.38 Å^{-1}. This suggests a bulk-like character of these states, as different values of $k_\perp$ are probed in different parts of the image. A bulk-like character is also consistent with calculations of the projected band structure shown in Fig. 2(d). Split-off surface states on or near the extrema of bulk bands are frequently encountered on Bi, Sb and $\mathrm{Bi}_{0.9}\mathrm{Sb}_{0.1}$ surfaces [8, 13, 18].

Figure 2(c) shows a similar cut through only one of the states, taken along the blue line in Fig. 2(a) but with a lower photon energy and higher energy and k-resolution (25 meV and 7 mÅ^{-1}). The state appears as a single feature on top of the Λ-shaped bulk bands. It can be estimated to have a highest binding energy of ≈ 150 meV. The full width at half maximum of the state at E_F (in k) is found to be ≈ 82 mÅ^{-1}, which is somewhat broader than for other Bi, Sb and $\mathrm{Bi}_{0.9}\mathrm{Sb}_{0.1}$ surfaces and for most other quasi one-dimensional systems with character [19–22]. Such broadening can be due to interaction with bulk states, as found on other bismuth surfaces [10, 13]. As shown in Fig. 2(d) and (e), the surface state is always quite close to, and partly in, the bulk band continuum, especially if the (1×2) reconstruction leads to an appreciable back-folding of the bulk bands.

It can be expected that the broken inversion symmetry at the surface leads to a strong spin-splitting of the surface states as on all other Bi surfaces studied so far [13, 24]. This splitting is not directly visible in the surface state band but should be detectable as a spin-asymmetry in spin- and angle-resolved photoemission.

Figure 3 shows the result of such an experiment in which the spin-dependent photoemission intensity was measured for a scan of the azimuthal angle crossing the surface state Fermi contour at an off-normal angle of 35° (see Fig. 3(b)). The spin integrated intensity in Fig. 3(a) shows a single peak, while the spin polarization data (Fig. 3(c)) have a pronounced left-right asymmetry, revealing the presence of two unresolved spin-split bands. Further analysis was based on a two-step method described in detail elsewhere [25]. In short, the spin-integrated intensity of Fig. 3(a) was first fitted by two Gaussian peaks to quantify the relative contribution of the sub-bands. The

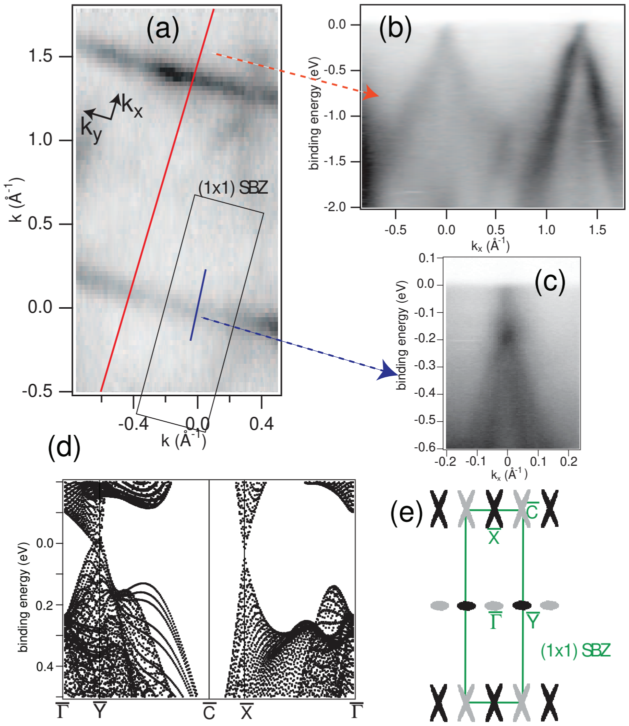

FIG. 2: (Color online) (a) Photoemission intensity at the Fermi energy (taken at a photon energy of $h\nu = 70$ eV), revealing the existence of a quasi one-dimensional Fermi line, passing through the origin. Also indicated is the surface Brillouin zone (SBZ) for the truncated bulk surface. (b) Energy vs. k cut through the data set of (a) along the red line in (a). (c) Cut along the blue line in (a) taken at $h\nu = 20$ eV. (d) Projected band structure along several high-symmetry directions in the truncated bulk SBZ. (e) Projection of the bulk Fermi surface on the (114) truncated bulk face. Regions marked in black represent states within 40 meV of the Fermi energy. Grey regions are the Fermi surface segments translated by the reciprocal lattice of the (1 × 2) reconstruction. The projections have been calculated using the tight-binding parameters of Liu and Allen [23].

separation between the peaks corresponds to 67 mÅ^{-1}. In a second step, the measured spin polarization curves were fitted by varying the polarization direction and its magnitude for each band, as shown in Fig. 3(c). The final result of the analysis is the degree of spin polarization for each band, as well as the orientation of the polarization vector.

The resulting spin polarization vectors are shown in Fig. 3(d). The states are found to be 100% spin-polarized. The polarization vector is inclined at 30±3° to the surface plane and is close to the $\bar{\Gamma}-\bar{Y}$ direction in the surface plane (angles of 10±3° and 5±3° for the two components). Similar data sets as the one shown in Fig. 3 have been taken by cutting through the same Fermi contour segment but using different off-normal emission angles. No appreciable difference in the result was found. Note that these experimentally determined spin directions are consistent with the general requirement of time-reversal symmetry, greatly increasing our confidence in the assumption that the surface state Fermi contour actually consists of two closely separated parallel lines which cannot be resolved in a spin-integrated experiment.

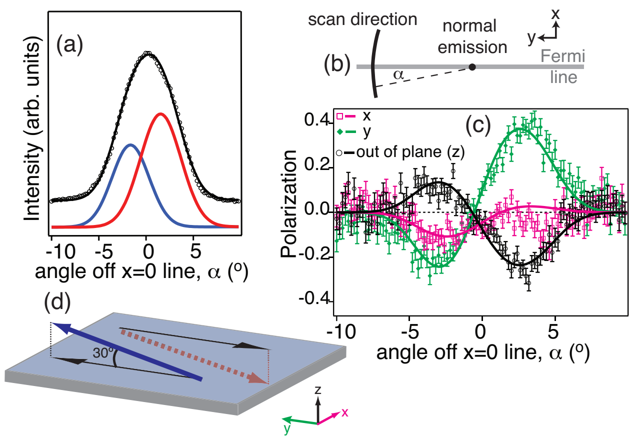

FIG. 3: (Color online) (a) Black open markers: spin-integrated photoemission intensity for an azimuthal angle scan through the surface Fermi line, taken at a polar off-normal emission angle of 35°. Two Gaussian components (red and blue) are fitted to the data to represent the two spin-split components. The black line is the result of the fit. (b) Sketch of the scan geometry. (c) Measured (markers) and fitted (lines) spin polarization data along the scan, split up into three mutually perpendicular components: x, y and out of the surface plane (z). (d) The directions of the experimentally determined spin polarization vectors relative to the surface plane.

Figure 4 summarizes the electronic structure of Bi(114), including the spin structure. Figure 4(a) shows a sketch of the surface Fermi contour with the main in-plane spin direction marked by arrows. The most important result is that the Fermi line is actually a Kramers-pair of spin-polarized states. The states are mainly polarized in the surface plane, as for a two-dimensional electron gas. The direction of the in-plane polarization, however, is fundamentally different: for a two-dimensional electron gas the spin polarization is *always perpendicular to the* **k***-vector* of the electrons. Here it is not: scans taken at different off-normal emission angles suggest that the spin is *always parallel to the direction of the Fermi contour*, independent of the position on the Fermi contour. This experimental finding, together with the straight Fermi lines, strongly support the notion that these states have one-dimensional character.

Figure 4(b) gives a qualitative picture of the possible surface state dispersion in the x direction, explaining the origin of the surface Fermi contour. For $k_x = 0$ the state is degenerate, as required by symmetry but a strong splitting is observed in the dispersion away from this line. Two bands cross E_F for very small values of k_x and give rise to the observed double Fermi contour. The other two

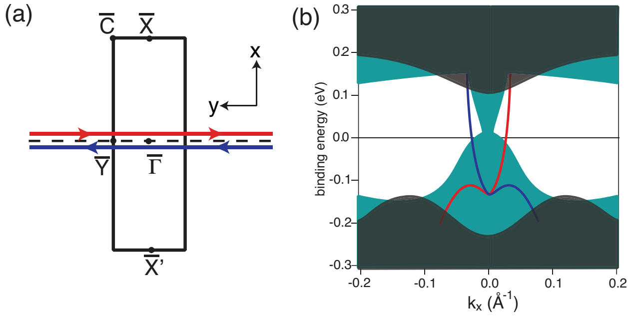

FIG. 4: (Color online) (a) Surface Fermi contour of Bi(114) resulting from this work. The arrows mark the main spin direction in the plane of the surface. (b) Qualitative sketch of the surface state dispersion in the x direction on top of the projected bulk band structure for the truncated bulk surface along $\bar{\Gamma} - \bar{X}$ (black area) and along $\bar{Y} - \bar{C}$ (green area).

bands are likely to disperse to higher binding energies and lose their surface character in the bulk continuum. This situation is very similar to the bands forming the electron pocket around the $\bar{\Gamma}$ point of Bi(111) [24]. Unlike on Bi(111), the bands forming the Fermi contour do not re-enter the occupied states since no further Fermi level crossings are observed in the first SBZ. Hence, they lose their surface character in the unoccupied bulk continuum.

Figure 4(b) also shows the calculated bulk band projection in the $\bar{\Gamma}-\bar{X}$ and the $\bar{Y}-\bar{C}$ directions of the truncated bulk SBZ to illustrate the two possible extreme cases of bulk band dispersion in the region of the surface state. As stated above, the surface state is degenerate with bulk states over a wide range of $\mathbf{k}$ and has only surface resonance character there, significantly contributing to its linewidth. The (1×2) reconstruction can further contribute to this by the back-folding of bands as indicated in Fig. 2(e).

It is interesting to compare our findings to the recent results on $\mathrm{Bi_{0.9}Sb_{0.1}}$(111) [8, 26] and to the more general findings associated with the quantum spin Hall effect [3–5, 27]. While pure Bi is not an insulator, as opposed to $\mathrm{Bi_{0.9}Sb_{0,1}}$, the electronic structures of the corresponding (114) surfaces can be expected to be similar. The twin Fermi line of Fig. 4(a) has a special topological stability; it shows exactly one Fermi level crossing on a line connecting two points of time-reversal invariant momenta ($\bar{\Gamma}$ and $\bar{X}$ in Fig. 4(a)) and is thus non-trivial. Correspondingly these states could be termed a 'topological metal'. The surface states should be better defined on $\mathrm{Bi_{0.9}Sb_{0.1}}$(114) because the continuum of underlying bulk states is removed. Moreover, the states bear a strong resemblance to the edge states in the quantum spin Hall effect [2] but they are readily one-dimensional by way of surface design and one can therefore expect intriguing transport properties.

In conclusion, we have shown that the (114) vicinal surface of Bi supports a one-dimensional non-degenerate surface state which gives rise to a Fermi contour consisting of two narrowly separated parallel lines. Bismuth is not an insulator so this surface state lies very close to and partially inside the bulk continuum. It is argued that the same surface on a topological insulator, e.g. $\mathrm{Bi_{0.9}Sb_{0.1}}$(114) could be a realization of the one-dimensional quantum spin Hall phase.

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