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Proving nontrivial topology of pure bismuth by quantum confinement

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The topology of pure Bi has been controversial because of its very small (∼10 meV) band gap. Here we performed high-resolution angle-resolved photoelectron spectroscopy (ARPES) measurements systematically on 14–202 bilayers Bi films. Using high-quality films, we succeeded in observing quantized bulk bands with energy separations down to ∼10 meV. Detailed analyses on the phase shift of the confined wave functions precisely determined the surface and bulk electronic structures, which unambiguously show nontrivial topology. The present results not only prove the fundamental property of Bi but also introduce a capability of the quantum-confinement approach.

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Semitmetal bismuth (Bi) has been providing an irreplaceable playground in condensed matter physics. Its extreme properties originating from the three-dimensional Dirac dispersion enabled the first observations of several important phenomena such as diamagnetism [1] and the various effects associated with Seebeck [2], Nernst [3], Shubnikov de Haas [4] and de Haas van Alphen [5]. Even now, numbers of novel quantum phenomena have been intensively reported on this system [6–13]. In spite of the enormous amount of research, one fundamental property of Bi has been controversial: its electronic state (QWS) subbands are formed inside bulk bands with inherent spin splitting. Despite the essential role in topological studies, a pure Bi crystal itself had long been believed topologically trivial based on several calculations [14–19], which had been considered to agree with transport [20] and angle-resolved photoelectron spectroscopy (ARPES) measurements [21–28]. However, a recent high-resolution ARPES result suggests the surface bands are actually different from previously calculated ones and Bi possesses a nontrivial topology [29, 30]. New transport measurements also imply the presence of topologically protected surface states [31, 32].

Nevertheless, the recent ARPES result has not yet been conclusive because it lacks clear peaks of bulk bands [29, 30]. In principle, surface-normal bulk dispersions can be measured by changing the incident photon energy, where the momentum resolution is determined from the uncertainty relation $\Delta z \cdot \Delta k_z \geq 1/2$ (Ref. [33]). ($\Delta z$ is an escape depth of photoelectrons.) However, the Dirac dispersion of Bi is so sharp against this resolution that h($\nu$)-dependent spectra show no clear peak [29, 30]. This is a serious problem because Bi has a very small (∼10–20 meV) band gap and a slight energy shift in bulk bands can easily transform a nontrivial case [Fig. 1(d)] into a trivial case [Fig. 1(e)]. In short, to unambiguously identify the topology of Bi, one must precisely determine both the surface and bulk electronic structures. One promising approach is using a thin film geometry, where quantum-well state (QWS) subbands are formed inside bulk band projections [31, 32]. Although QWSs originate from bulk states, they possess a two-dimensional character and can be clearly observed in ARPES measurements.

In this letter, we performed high-resolution ARPES measurements on Bi(111) films with thicknesses increasing from 14 to 202 BL (bilayer; 1 BL = 3.92 Å [20]). High-quality films enabled us to clearly observe the QWS subbands with energy separations down to ∼10 meV. After we confirmed the interaction between the top and bottom surface states in the 14 BL film, we systematically followed the evolution of the electronic structures. Detailed analyses on the phase shift of the QWS wave functions precisely determined the surface and bulk band dispersions. The revealed electronic structures unambiguously show that a pure Bi crystal has a nontrivial topology. The present results not only prove the fundamental property of Bi, but also highlight the QWS approach as a powerful tool to determine fine electronic structures.

A surface of a p-type Ge wafer cut in the [111] direction was cleaned in ultrahigh vacuum by several cycles of Ar$^+$ bombardment and annealing up to 900 K. Bi was deposited at room temperature and annealed at 400 K [30].
Although there is slight discrepancy in energy positions, structures obtained by the first-principles calculations. Two surface bands exist inside a band gap at TRIM without violating Kramers’ theorem. Therefore, in addition to the nontrivial scenario that SS1 connects to the conduction band (CB) at M [Fig. 2(e)], it is also possible that SS1 connects to VB in the bulk limit but that it is pushed into a gap in an ultrathin film by the surface-surface interaction [Fig. 2(f)]. Although Fig. 2(e) and (f) depict SS2 hybridizing with VB at M as suggested by previous studies, the overall structures show good qualitative agreement.

First we organize information regarding Bi topology. For the (111) surface of Bi, two spin-splitting surface bands, SS1 and SS2, bridge Γ and M points. Although experimental and theoretical results agree that both bands connect to the valence band (VB) around Γ point, a discrepancy lies in their connection around M point [29, 35, 38]. Based on Kramers’ theorem, a spin-splitting band cannot exist at time-reversal-invariant momenta (TRIM) [14, 21, 27–29]. Therefore we can limit the possible cases to those depicted in Fig. 1(d)-(g). We note a nontrivial topology exists only in Fig. 1(d) case, which is distinguished from the other cases in that SS1 and SS2 bands are non-degenerate at M.

We start from an observation of an ultrathin film. Figure 2(a) shows the Fermi surface of a 14 BL Bi(111) film measured at $h\nu = 21$ eV. The shape is very close to that of bulk Bi [27, 29, 44]. Figure 2(b) shows the corresponding band structures along ΓM direction with calculated bulk projections. Two surface bands exist inside the bulk band gap and QWS subbands inside the bulk projection. The observed bands are consistent with previous reports [33, 35, 40]. Figure 2(c) illustrates band structures obtained by the first-principles calculations. Although there is slight discrepancy in energy positions, the overall structures show good qualitative agreement. It is clear that SS1 and SS2 bands are non-degenerate at M, which appears to suggest Bi is topologically nontrivial based on Fig. 1(d)-(g). However, in an ultrathin Bi film whose thickness is as small as a decay length of the surface state, the top and bottom surface states can interact with each other and modify their shape from such a surface-surface interaction. These merged states possess even numbers of electrons and can exist inside a band gap at TRIM without violating Kramers’ theorem. Therefore, in addition to the nontrivial scenario that SS1 connects to the conduction band (CB) at M [Fig. 2(e)], it is also possible that SS1 connects to VB in the bulk limit but that it is pushed into a gap in an ultrathin film by the surface-surface interaction [Fig. 2(f)]. Although Fig. 2(e) and (f) depict SS2 hybridizing with VB at M as suggested by previous studies, the overall structures show good qualitative agreement.

The pressure was kept at $\sim 1 \times 10^{-8}$ Pa during the deposition. The film thickness was precisely measured with a quartz thickness monitor. Qualities of the substrate and the film were confirmed from low-energy electron diffraction measurements. ARPES measurements were performed at BL-9A of HiSOR and BL-21B1 of NSRRC. In BL-9A a high-intensity unpolarized Xe plasma discharge lamp (8.437 eV) was used in addition to synchrotron radiation (21 eV). The measurement temperature was kept at 10 K and the total energy resolution was 12 meV for 21 eV. Solid lines in (b) indicate bulk projections calculated by a tight-binding method [20]. (c) Band structures obtained by the first-principles calculations for a 14 BL Bi slab. (d) Plane-averaged electronic charge densities within the film calculated at the four $k$ points marked in (c). (e), (f) Possible band assignments in an ultrathin Bi film. Grey areas illustrate positions of the VB maximum (VBM) and CB minimum (CBM).
Figure 4(b) shows $E-k_{\perp}$ dispersion obtained using this relation (39). The error bars are estimated by uncertainties in thicknesses and fitted peak positions. Here the surface-normal direction at $\bar{M}$ corresponds to $LX$ [Fig. 1(a)] and Bi has its Dirac dispersion along this direction. Figure 4(c) shows the tight-binding result (20). The experimental data are indeed perfectly fitted by the solid line in Fig. 4(b); the fitted result is 

$$E = \alpha k_{\perp}^\text{exp} + \beta$$

where $\alpha = 3.58 \pm 0.01$ eV.Å and $\beta = 0.024 \pm 0.002$ eV.

To experimentally extract information concerning $k_{\perp}$, we note that some QWSs have the same binding energy but different $N$ and $n$. Since the phase shift $\Phi$ can be regarded as only a function of $E$ (34), we can derive

$$k_{\perp, \text{exp}} = \frac{\pi}{t} \left( n - n' \right)$$

Figure 4(b) shows $E-k_{\perp, \text{exp}}$ dispersion obtained using this relation (39). The error bars are estimated in thicknesses and fitted peak positions. Here the surface-normal direction at $\bar{M}$ corresponds to $LX$ [Fig. 1(a)] and Bi has its Dirac dispersion along this direction. Figure 4(c) shows the tight-binding result (20). The experimental data are indeed perfectly fitted by the solid line in Fig. 4(b); the fitted result is $E = \alpha k_{\perp, \text{exp}} + \beta$, where $\alpha = 3.58 \pm 0.01$ eV.Å and $\beta = 0.024 \pm 0.002$ eV.

To test this hypothesis, we analyzed the QWS energy positions in more detail. Figure 4(a) shows the energy distribution curves (EDCs) extracted at $\bar{M}$ for each thickness. Peak positions were determined using Lorentzian fittings. These energy positions can be simply described using the phase accumulation model, which assumes electronic waves propagating forward and backward across the film and being reflected at the top and bottom surfaces. The model provides the expression

$$2k_{\perp}(E)N(E)t + \Phi(E) = 2\pi(n-1)$$

The first term represents the phase shifts in propagation, with $k_{\perp}(E)$ and $N(E)$ denoting the surface-normal dispersion and the number of bilayers, respectively, and $t$ the thickness of one bilayer (3.92 Å (20)). $\Phi(E)$ is the total phase shift at the top and bottom surfaces and $n$ is a quantization number.

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Now that we have experimentally obtained $k_{\perp}(E)$, we can derive a total phase shift using equation (1). For this purpose, we used $n = 1$ and $n = 2$ QWS energy positions and corresponding thicknesses. The result shown in Fig. 4(d) exhibits an almost constant relation in this energy range. The fitted value by a constant function is $\Phi_{\exp} = (-1.70 \pm 0.03)\pi$, which is similar to those reported in ultrathin Bi films on a Si substrate [45]. Furthermore, we compared the experimental and analytical results by plotting $N$ against $E$ (a structure plot) in Fig. 4(c). The latter is obtained using

$$N(E) = \frac{2\pi(n-1) - \Phi_{\exp}}{2k_{\perp,\exp}(E)t}$$

(3)

It excellently reproduces the experimental data not only for $n = 1$ and $n = 2$ QWSs but also for each of the other $n$ values. The consistency of the entire analysis shows that SS2 band around $\bar{M}$ indeed becomes a part of QWSs, and also demonstrates the validity of the obtained phase shift.

As a final step we follow the evolution of VB and SS1 bands at $\bar{M}$ to identify Bi topology. Figure 5(a) shows EDCs magnified around a peak near $E_F$. The peak broadens as thickness increases and finally exhibits multiple peaks. This is attributed to a tail of a QWS located above $E_F$. We noted the clear threshold between 43 and 58 BL [39]. Extracted peak positions were plotted against an inverse thickness $1/N$ along with VBM ($n = 1$ QWS) peaks in Fig. 5(b). Using equation (1), an exponential decay against the film thickness [50] but the linear dispersion shown in Fig. 4(c). A possible reason is a strain effect from the Ge substrate. However, this can be excluded by considering the $1/N$ dependence. A lattice strain exhibits an exponential decay against the film thickness [50] but the linear dispersion in Fig. 4(b) does not appear to fit an exponential decay. Moreover, an exponential function has downward convexity with $1/N$, which further reduces the possibility that VBM and CBM cross each other.

In conclusion, we were able to unambiguously prove that pure Bi is topologically nontrivial. Although the interaction between the top and bottom surface states does exist as revealed by calculations, the splitting between SS1 and SS2 is not a consequence of the interaction but rather the electronic structure unique to Bi. The present result provides an important insight in recent attempts to detect novel quantum phenomena on pure Bi, where the three-dimensional massive Dirac fermion and its nontrivial topology can show an interesting connection. Furthermore, the topologically protected surface states with a giant spin splitting offer great potential in spintronics applications. Recent transport measurements have shown Bi keeps its unique surface transport at ambient pressure [31, 32]. A possible application of Bi surface states to valleytronics was also recently reported [13].
Finally we also emphasize the capability of the QWS approach we used. Further advancing the established method, we demonstrated that systematic analyses on QWSs can precisely assign and map surface and bulk bands even at ∼10 meV scale and can reveal hybridizations between them. Novel topological materials recently predicted can have as small energy scales as observed here in Bi. Precise determination of surface and bulk electronic structures is indispensable in driving forward topological studies, where the present method can be one of the most powerful tools.

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