Interfacing 2D and 3D Topological Insulators: Bi(111) Bilayer on Bi_2Te_3

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We report the formation of a bilayer Bi(111) ultrathin film, which is theoretically predicted to be in a two-dimensional quantum spin Hall state, on a Bi_2Te_3 substrate. From angle-resolved photoemission spectroscopy measurements and *ab initio* calculations, the electronic structure of the system can be understood as an overlap of the band dispersions of bilayer Bi and Bi_2Te_3. Our results show that the Dirac cone is actually robust against nonmagnetic perturbations and imply a unique situation where the topologically protected one- and two-dimensional edge states are coexisting at the surface.

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Topological insulators, realized in materials with strong spin-orbit interaction, are gaining increasing attention in condensed matter physics. Mathematically characterized by the Z_2 topological number, they are band insulators but have metallic edge (surface) modes that are protected by time-reversal symmetry. It is predicted that they may be used in application for spintronics or quantum computing [1].

The presence of such systems has been well established experimentally by measuring the helical Dirac-like surface-state band dispersion for various Bi-based materials with (spin- and) angle-resolved photoemission spectroscopy (ARPES) [2–4] for three-dimensional (3D) materials. Theoretically, the topological surface states are described as ''domain-wall'' states at the interface of two systems that have different topological properties (a sign change of the Dirac mass) [1]. Their existence is always guaranteed at the topological-trivial interface. To investigate how the Dirac cone is affected by perturbations, theorists have calculated the electronic structure for systems where the whole surface is terminated with dielectrics [5] or surfaces that have different terminations [6]. In terms of experiment, an apparent ''gap opening'' of the Dirac cones was found by nonmagnetic impurity adsorption possibly due to band bending effects [7,8].

For two-dimensional quantum spin Hall states (2D QSH), there has been only one case where elaborated work has been performed: HgTe/CdTe quantum wells [9]. Although the topological protection is more complete in the one-dimensional (1D) edge states compared to the 2D surface states of 3D topological insulators, only a few works have been done experimentally [10]. An interesting system predicted to be in a 2D QSH phase is a single-bilayer Bi(111) ultrathin film, which is said to have much shorter edge-state penetration length scales compared to HgTe/CdTe quantum wells [11,12]. But since it is the thinnest limit for a 2D system similar to graphene, it has not been realized experimentally. One reason is that Bi prefers the black-phosphorous structure instead of the rhombohedral structure for atomic-layer thicknesses [13].

In the present Letter, we show that we succeeded in fabricating a single bilayer of Bi(111) on Bi_2Te_3(111) since they both form in layers with a hexagonal lattice [Figs. 1(a) and 1(b)] and investigated how the Dirac cone is affected by this Bi termination. From ARPES and first principles calculations, we show for the first time that the Dirac cone of Bi_2Te_3 is actually robust against nonmagnetic perturbations. The present system is ideal to study the 1D edge states of a 2D QSH Bi bilayer as well as to explore the novel spin transport phenomena due to the interplay of 1D edge and 2D surface states.

The ARPES experiments were performed at BL-5U of UVSOR using the MBS-Toyama A-1 analyzer at 10 K. The energy and the angular resolutions were 20 meV and 0.2°, respectively. The calculations have been performed by the full-potential linearized augmented plane wave method in film geometry as implemented in the FLEUR program [14], and the generalized gradient approximation [15] has been used for the description of the exchange-correlation potential.

All the film fabrication and measurements were done *in situ*. First, a clean Si(111)-(7 × 7) surface was prepared on an *n*-type substrate (P-doped, 1–10 Ω · cm at room temperature) by a cycle of resistive heat treatments. Then Bi was deposited on the 7 × 7 structure at ~400 K under Te-rich conditions. Such a procedure is reported to result in a quintuple-layer-by-quintuple-layer [(QL), 1 QL = 10.2 Å, Fig. 1(f)] epitaxial film formation [16]. This can be seen in the clear reflection high-energy electron diffraction (RHEED) pattern in Fig. 1(e), indicating the 1 × 1 periodicity of the Bi_2Te_3(111) surface as well as the Kikuchi pattern. It is also known that high-quality epitaxial Bi(111) films can be formed on Si(111)-(7 × 7) in bilayers [Fig. 1(c)] [(BL), 1 BL = 3.9 Å] [13]. Theory predicts that

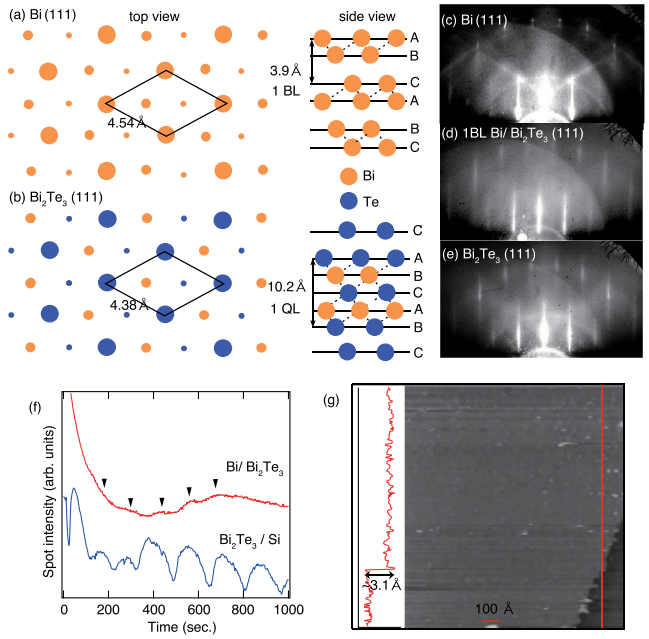

FIG. 1 (color online). (a),(b) Crystal structure of Bi(111) (a) and Bi_2Te_3(111) (b), respectively. The top and side views are shown explicitly with the unit cells. (c)–(e) RHEED patterns of a Bi(111) surface (c), a single-bilayer Bi(111) grown on Bi_2Te_3(111) (d), and Bi_2Te_3(111) (e), respectively. The electrons are incident along the $[11\bar{2}]$ direction of the underlying Si substrate, and the energy is 15 keV. (f) RHEED spot intensity of the (00) spot during Bi and Bi_2Te_3 film growth showing clear oscillation and the periodicity. (g) A STM image of the single-bilayer Bi(111) film ($V_{tip} = -3$ V, $I = 0.35$ nA). 3.1 Å corresponds to the step height of Si(111).

a single-BL Bi(111) is in a topological nontrivial state [11,12]. However, it has not been experimentally demonstrated, because the minimum thickness for Bi(111) formed on Si(111) is 6 BL due to the smaller cohesive energy for the Bi{012} phase at small thicknesses [13]. Bismuth and Bi_2Te_3 are layered materials with similar crystal structure, and the lattice constant is nearly the same [Figs. 1(a) and 1(b)]. Therefore, we have deposited an additional Bi bilayer on the Bi_2Te_3(111) surface to see if the realization of the bilayer Bi(111) film is possible. Figure 1(d) shows the RHEED pattern of such a structure. It shows little change from that of the pristine Bi_2Te_3 [Fig. 1(e) with the clear 1×1 pattern], meaning the successful formation of a bilayer Bi(111). The STM image also shows a flat film structure [Fig. 1(g)]. Figure 1(f) shows the RHEED spot intensity profile during Bi growth which shows the bilayer-by-bilayer growth mode. We have carefully checked the oscillation period and calibrated the Bi film coverage, which was also cross-checked by the formation of a Si(111)$\beta\sqrt{3} \times \sqrt{3} -$ Bi surface superstructure as well as the allotropic transformation at 6 BL [13].

In order to gain insight into how the Dirac-surface states are affected by the Bi adsorption and the band dispersion of bilayer Bi, we have measured the electronic structure with ARPES. Figures 2(a) and 2(b) show the band dispersion along the $\bar{\Gamma}$-$\bar{K}$ direction for the 18 QL thick pristine Bi_2Te_3 (a) and its momentum distribution curves (MDC) (b), respectively. The state with strong intensity below the Fermi level (E_F) near the $\bar{\Gamma}$ point is the bulk (film) conduction band as it changes with the photon energy [Fig. 2(e)]. The feature with linear band dispersion beside it corresponds to the Dirac-cone-type surface state. An important aspect of the Dirac cone in Bi_2Te_3 is that the Dirac point overlaps with the bulk valence band ["M" shape around 0.35 eV in Fig. 2(a)]. In the MDC, the Dirac point cannot be easily recognized due to the weak intensity, but it is speculated to be 0.37 eV below the Fermi level. The situation changes drastically upon Bi-bilayer adsorption. Figures 2(c) and 2(d) show the band dispersion along the $\bar{\Gamma}$-$\bar{K}$ direction for the single-BL Bi(111) terminated 18 QL thick Bi_2Te_3 (c) and its MDC (d), respectively. In addition to the features near the $\bar{\Gamma}$ point, we can see a pair of states dispersing upwards from $k \sim \pm 0.6$ Å^{-1} at 0.6 eV below E_F. Although the Dirac cone itself seems to be unaffected, there is a slight change. This can be seen more clearly in Fig. 2(d). At the $\bar{\Gamma}$ point near 0.3 eV, the linear dispersion is now explicitly seen. In contrast to what is shown in Fig. 2(b), the Dirac point in this case does not have any overlap with other states nearby. This can be seen even better when we change the probing photon energy to $h\nu =$ 30 eV, which enhances the photoemission intensity for the surface states and diminishes that of the bulk [Figs. 2(e) and 2(f)]. For the pristine Bi_2Te_3, the upper Dirac cone and the lower bulk band do not connect smoothly [Fig. 2(e)], whereas the connection between the upper and lower branches is smooth in the Bi-bilayer terminated Bi_2Te_3 [Fig. 2(f)].

The above result shows that, although two new bands appeared by Bi adsorption, the Dirac cone is only slightly modified. This is in agreement with the fact that the Dirac cone is topologically protected [17]. To verify what has actually changed and what has not, we have performed *ab initio* calculations. First, Fig. 3(a) shows the band dispersion of the freestanding Bi bilayer [18]. Bands a and b are two of the three doubly degenerate, occupied π bands dispersing upwards to the $\bar{\Gamma}$ point, and, from the parity analysis, this band structure was shown to possess a nontrivial Z_2 number [12]. However, the dispersion measured experimentally shown in Figs. 2(c) and 2(f) shows an electronic structure much more complicated and suggests that our Bi bilayer is not isolated from the underlying substrate. Figure 3(b) shows the dispersion for the 6 QL Bi_2Te_3 slab with the spin polarization (we show the spin component that is oriented in-plane perpendicular to k). It has been reported that 6 QL is sufficiently thick enough to reproduce the Dirac cone of Bi_2Te_3 [19] and indeed agrees with the experimental results of Figs. 2(a), 2(b), and 2(e).

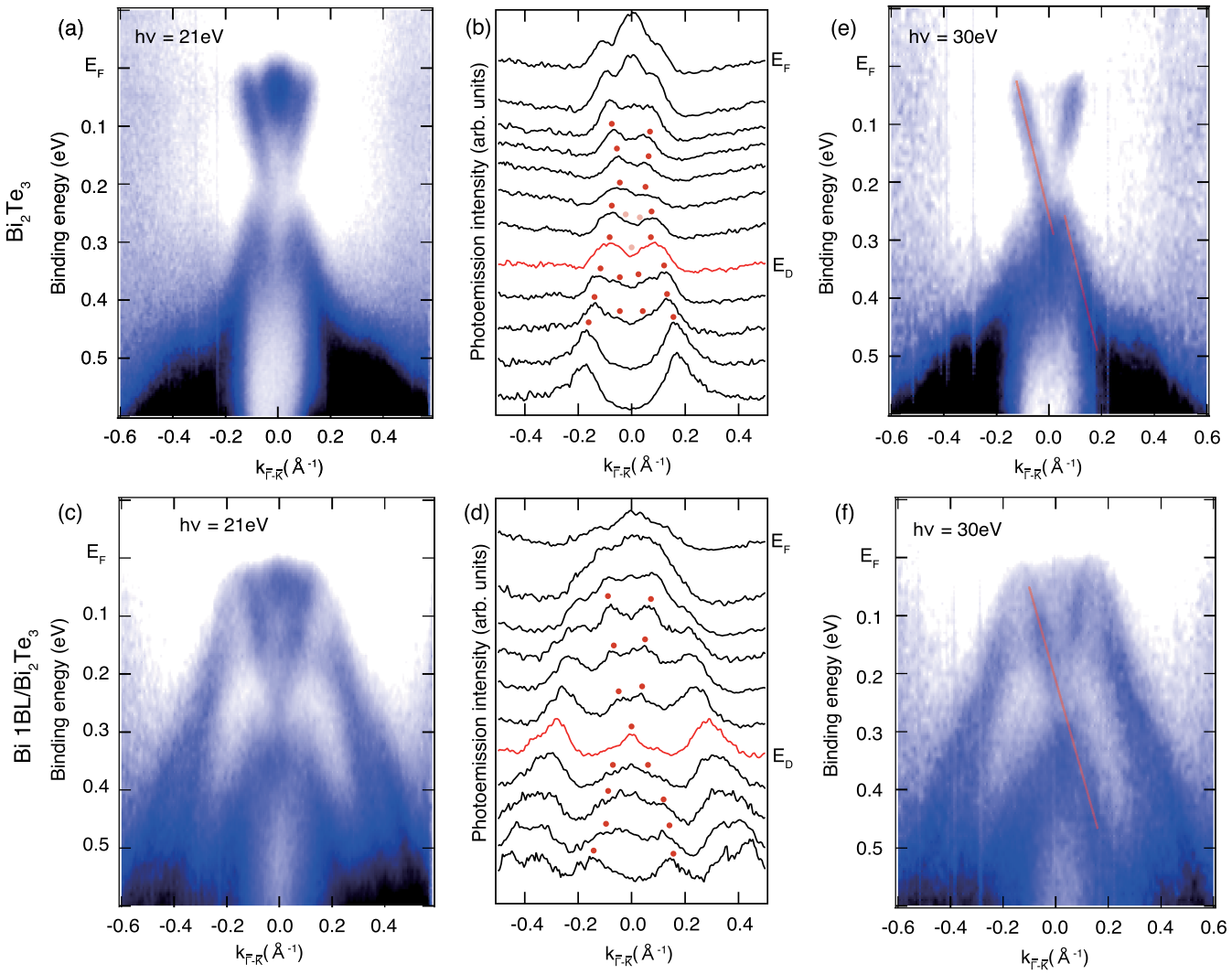

FIG. 2 (color online). (a)–(d) The band dispersion image along the $\bar{\Gamma}$-$\bar{K}$ direction taken at $h\nu = 21$ eV for a 18 QL thick Bi_2Te_3 (a) and its momentum distribution curves (b), respectively. The same for a single-bilayer Bi(111) terminated Bi_2Te_3 [(c) and (d)]. (e), (f) The band dispersion image along the $\bar{\Gamma}$-$\bar{K}$ direction taken at $h\nu = 30$ eV for a 18 QL thick Bi_2Te_3 (e) and single-bilayer Bi(111) terminated Bi_2Te_3 (f), respectively.

Finally, we have calculated the dispersion for 6 QL Bi_2Te_3 terminated with a single-BL Bi [Fig. 3(c)] to take into account the Bi/Bi_2Te_3 interaction. The lattice constant for the Bi(111) bilayer was taken as 4.38 Å, which is the value for Bi_2Te_3(111) [Fig. 1(b)]. Relaxation was allowed in the direction perpendicular to the surface. The agreement between the experiment and calculation is excellent, as shown in Fig. 3(d) (the calculation has been shifted by 0.13 eV). Especially the features of the two upward-dispersing bands [marked by *a* in Fig. 3(c)] are reproduced nicely as well as the situation near the Dirac point. Indeed, Fig. 3(c) clearly shows that there is a well-isolated Dirac cone existing near the surface of a Bi BL/Bi_2Te_3 system, which is in contrast to that of the pristine Bi_2Te_3 [Fig. 3(b)]. We also notice that the Dirac cone maintains the spin helical structure [Figs. 3(c), 3(e), and 3(f)]. Thus we found that, upon Bi adsorption, the surface states are robust but the Dirac point becomes well isolated from the bulk. Furthermore, if we look into the band structure more carefully, we also realize that the bands *a* themselves resemble Dirac cones and connect the valence and conduction bands [Figs. 3(e) and 3(f)]. Therefore, in addition to Dirac cone *i*, two other conelike structures appear (*ii* and *iii*). If the Fermi level can be tuned near Dirac point *i*, only the three Dirac states will cross the Fermi level in the $\bar{\Gamma}$-$\bar{M}$ direction [Fig. 3(e)], representing the topological protection.

Despite the interaction at the interface, we would like to note that the additional bands that appeared by Bi adsorption [Fig. 3(f)] can be naturally understood by considering that of a pristine Bi(111) bilayer [Fig. 3(a)]. The upward-dispersing band pair is a result of splitting of the

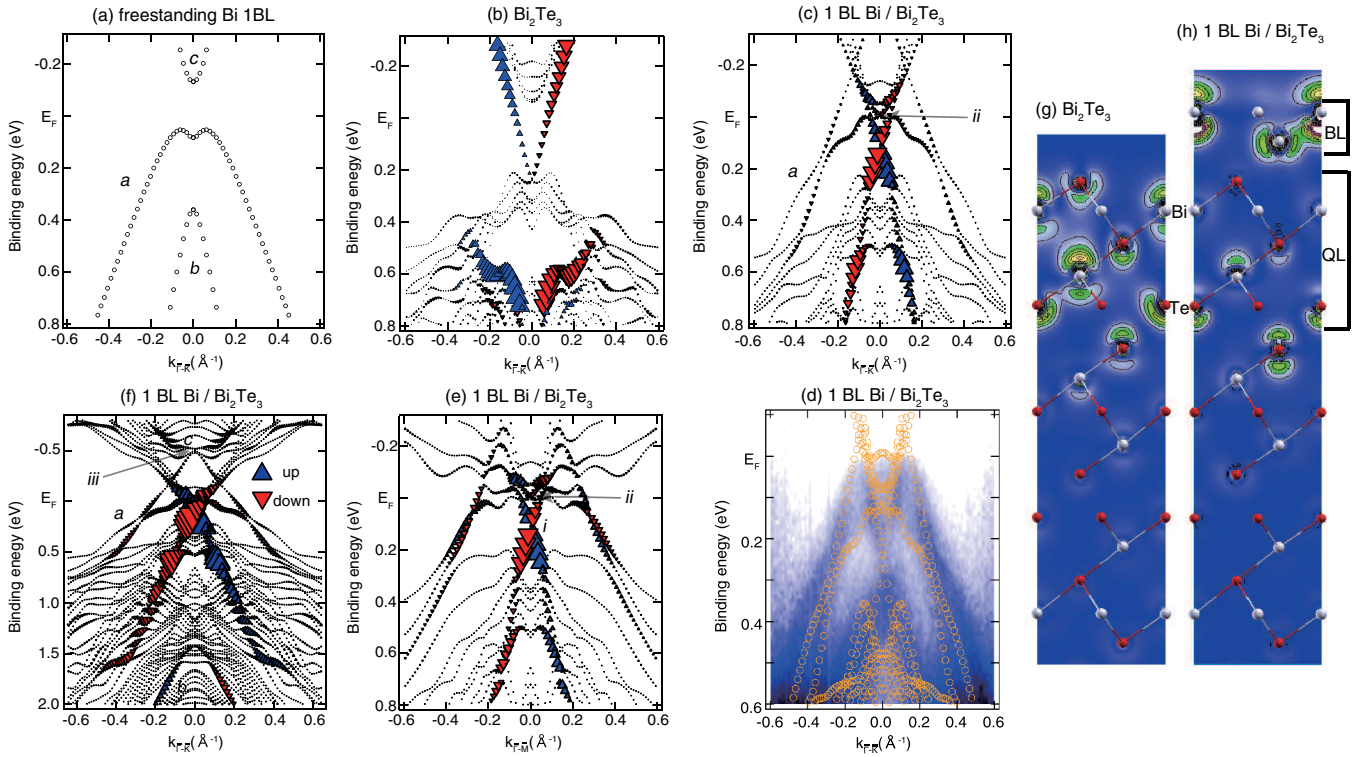

FIG. 3 (color online). (a)–(c) Calculated band structure along the $\bar{\Gamma}$-$\bar{K}$ direction for a freestanding Bi bilayer (a), 6 QL thick Bi_2Te_3 (b), and single-bilayer Bi(111) terminated Bi_2Te_3 (c), respectively. (b) and (c) also show the spin polarization. (d) The calculated band structure of (c) overlapped on the experimentally obtained band dispersion of Fig. 2(c). (e) The spin-polarized band structure of single-bilayer Bi(111) terminated Bi_2Te_3 along the $\bar{\Gamma}$-$\bar{M}$ direction. (f) Spin-polarized band dispersion of (c) shown in a larger energy range. The states marked *a*, *b*, and *c* are the counterparts of the freestanding Bi bilayer shown in (a). The symbols *i*, *ii*, and *iii* show the Dirac points. The spin component shown is oriented in-plane perpendicular to *k*. (g),(h) Spatial distribution of the charge density of the surface state at the Dirac point on the Bi_2Te_3(111) surface (g), and the single-bilayer Bi(111) terminated Bi_2Te_3 surface (h), respectively.

spin-degenerate band of Bi(111) bilayer due to symmetry breaking at the interface (marked *a*). Bands *b* and *c* also have counterparts, which are shifted a little bit in energy [20]. Thus, except for some hybridization effect, the original bands of Bi(111) bilayer and the Dirac cone of Bi_2Te_3(111) persist even for the present system. This shows that the band structure of the deposited Bi(111) BL is still compatible with the 2D QSH state as predicted by theory [11,12].

One thing that has actually changed by Bi adsorption is the charge distribution at the Dirac point. Figures 3(g) and 3(h) show the results for the pristine Bi_2Te_3 (a) and Bi-terminated Bi_2Te_3 (b), respectively. For Bi_2Te_3, the charge is mostly localized in the first QL. Although they are termed as surface states, they extend 10 Å into the interior crystal. This has been shown in a previous report [6]. On the contrary, the distribution in Fig. 3(h) shows that the charge is mostly distributed at the topmost Bi bilayer and extends into the vacuum. This shows that, although the presence of the Dirac cone itself is robust against Bi adsorption due to the different topological numbers, the charge becomes localized at the surface and becomes more like a conventional Shockley state [21].

By tuning the Bi coverage slightly from 1 BL, it should be possible to fabricate Bi islands or nanoribbons of bilayer height with some bare Bi_2Te_3. It has been shown that, at the edges of such Bi honeycomb systems, topological metallic edge states are present irrespective of their geometry (armchair or zigzag [12]). The present films would be indeed a nice system to observe such edge states directly with scanning tunneling microscopy or spectroscopy measurements. Furthermore, a unique situation is realized where a 1D topological edge state is coexisting with 2D topological surface states. By combining the two, a ballistic transport of 2D surface states mediated by 1D edge states may be possible, leading to a novel spin channel at the surface. It will also be interesting to investigate how two 1D edge states from different nanoribbons interact with the aid of the 2D surface states.

In conclusion, we found that a single-bilayer Bi(111) ultrathin film can be experimentally realized on a Bi_2Te_3 substrate and has similar band dispersions to the calculation for the QSH state. Furthermore, we found that the original Dirac-cone surface states of the substrate Bi_2Te_3 is robust against Bi adsorption, experimentally demonstrating the ''topological protection.'' The present system is an

ideal one to study the 1D edge states as well as to explore the intriguing phenomena associated with the coexistence of 1D and 2D edge states.

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[17] One may argue that since bilayer Bi(111) is in a 2D QSH state, the Dirac cone should vanish since both bilayer Bi and Bi_2Te_3 are topologically nontrivial. But they have different dimensions, and, with respect to 3D Z_2 number, bilayer Bi is trivial. Therefore, the Dirac cone should be robust.
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[20] Although bands *a* and *c* come closer compared to the freestanding case, a gap on the order of 20 meV still exists at the $\bar{\Gamma}$ point. Furthermore, since the parity of bands *a* and *c* is the same, the topological properties do not change even if there was an inversion.
[21] We believe that there is hardly any band bending effect at the Bi/Bi_2Te_3 interface in this case, which is in contrast to Refs. [7,8]. This is probably because there should be no significant difference in the electronegativity of the topmost Bi bilayer and the Bi layer in Bi_2Te_3. Furthermore, the Debye length of the space-charge region is $\sim$20 nm, on the order of the Bi_2Te_3 film thickness. Therefore a band bending in the atomic-layer scale is not expected even considering the slight difference in the bulk doping.