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Citation: *Appl. Phys. Lett.* **98**, 222503 (2011); doi: 10.1063/1.3595309

View online: <http://dx.doi.org/10.1063/1.3595309>

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Robust surface electronic properties of topological insulators: Bi₂Te₃ films grown by molecular beam epitaxy

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(Received 8 April 2011; accepted 7 May 2011; published online 1 June 2011)

The surface electronic properties of the important topological insulator Bi₂Te₃ are shown to be robust under an extended surface preparation procedure, which includes exposure to atmosphere and subsequent cleaning and recrystallization by an optimized *in situ* sputter-anneal procedure under ultrahigh vacuum conditions. Clear Dirac-cone features are displayed in high-resolution angle-resolved photoemission spectra from the resulting samples, indicating remarkable insensitivity of the topological surface state to cleaning-induced surface roughness. © 2011 American Institute of Physics. [doi:10.1063/1.3595309]

Recently, a new class of materials, called the topological insulators (TIs), has been predicted theoretically and experimentally observed.¹ TIs are characterized by odd number of Dirac cones that show a linear energy dispersion similar to relativistic particles. Hence, carriers at the surface of TIs form a two-dimensional electron gas with unparalleled properties, such as extremely high mobilities or dissipationless spin-locked transport, which may lead to new applications in the field of spintronics or quantum computing. Bi₂Te₃ is a narrow gap semiconductor that has been traditionally investigated as a thermoelectric material, however, very recently the TI behavior has been observed at its surface.² To date, the studies of bismuth telluride's TI properties have mainly been carried out on bulk crystals prepared by the melt-growth or self-flux method,^{2,3} which result in heavily doped *n*-type material due to formation of defects. To compensate the *n*-type doping, the Bi₂Te₃ crystals are usually heavily doped with Sn or Ca, which, however, strongly degrades transport properties. Looking forward to future device applications, it is desirable to develop growth procedures for intrinsic *undoped* thin films of Bi₂Te₃, for example, by means of molecular beam epitaxy (MBE). Very recently Li *et al.*⁴ reported on MBE-growth of high quality single-crystal Bi₂Te₃ epilayers onto Si(111) substrates that showed the linear energy dispersion as determined by means of *in situ* angle-resolved photoelectron spectroscopy (ARPES). Implementing such films in device structures will necessarily require some postgrowth surface treatment or the formation of interfaces. As the appearance of topologically insulating behavior is tied to electronic states at the boundary of the sample, the question arises how stable these electronic properties are with respect to a subsequent surface modification.

In this letter, we report on ARPES measurements of MBE-grown Bi₂Te₃/Si(111) epilayers, which have been exposed to air prior to the measurements. We have observed a remarkable recovery of the TI behavior of the Bi₂Te₃ surface in the course of a standard cleaning procedure under UHV

conditions, which directly proves the insensitivity of the topological surface states to surface roughness and impurities, in agreement with recent scanning tunneling microscopy studies.^{5–8}

The Bi₂Te₃ epilayers were grown by means of MBE on *n*-type Si(111) wafer with details of the procedure explained in Ref. 9. The atomic force microscopy (AFM) images of as-grown films show atomically smooth surfaces with atomic steps of ≈ 1 nm height, related to the Bi₂Te₃ quintuple layer (QL). Pieces of approximately 10 mm \times 10 mm were cut out of the wafer and transferred into the high-resolution ARPES machine.¹⁰ The temperature of the sample during annealing was controlled by a calibrated infrared pyrometer through the standard UHV viewport. All data shown in this letter were measured using either nonmonochromatized He I ($h\nu=21.22$ eV) or monochromatized Kr ($h\nu=10.03$ eV) light.¹⁰ During the measurements, samples were mounted on the cryostat cooled either by liquid nitrogen or liquid helium, and the resolution of the spectrometer was set to 20 meV for all the presented spectra. The sample has been cleaned by cycles of Ar-ion sputtering¹¹ at 500 eV and subsequent annealing at 250°. The Auger electron spectroscopy (AES) results for one of the films are shown in Fig. 1. The appearance of the Si substrate peak in the AES spectra (inset in Fig. 1) allowed one to deduce a sputtering rate of ≈ 2.5 nm/min. This rate was also confirmed by x-ray reflectivity measurement on other samples taken out of vacuum after a few cleaning cycles. AFM measurements revealed that our cleaning procedure caused an increased surface roughness with an rms value of ≈ 1 nm.

Figures 2(a)–2(e) show the set of ARPES $E_B(k)$ maps reflecting the cleaning process of the sample, in which the thickness of the Bi₂Te₃ decreases with subsequent cleaning cycles. Top panels show the details of the photocurrent near the Fermi level, and bottom panels offer a wider overview, which allows comparison of the bulk and surface features. The most pronounced feature of the spectra is the Dirac-cone structure and sharp bulk dispersions, which compare well to the data from cleaved single crystals^{3,13,14} or films studied

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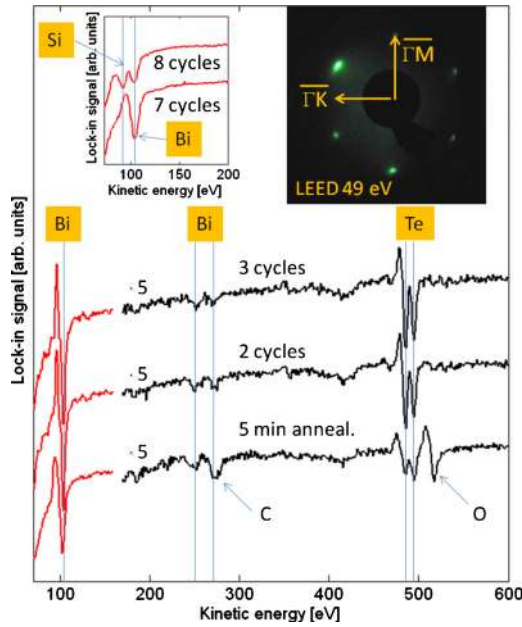


FIG. 1. (Color online) Auger spectra showing progress in cleaning of the sample. Each cleaning cycle consisted of 1 min 500 eV Ar-ion sputtering and 5 min annealing at 250 °C. LEED pattern after two cleaning cycles is also shown.

in situ.⁴ Furthermore, the top panels clearly show the bulk conduction band minimum (CBM) moving upward with decreasing thickness of the film, a process which is accompanied by the increasing valence band spectral weight in comparison to the Dirac-cone surface state. Strikingly, this clear movement of ≈ 100 meV between panels (a) and (c) is not accompanied by the related shift of the bulk bands (dashed line in these panels indicates unchanged binding energy of the most pronounced bulk feature), therefore it reflects changes in the electronic band structure more complex than

just a trivial uniform shift of all bands. For ultrathin films [panel (e)] a clear change in bulk bands is observed, which seems to indicate a collapse of the band structure properties, perhaps because in such thin films, the crystalline properties cannot be re-established after sputtering.

Figure 2(f) clearly reveals the complex hexagram shape of the Bi_2Te_3 Dirac-cone Fermi surface dispersion, as seen before by Chen *et al.*³ In the Fermi surface image there is no significant spectral weight related to the bulk conduction band at the zone center. In Fig. 2(h) the Kr-spectrum measured on the 11 QL thick film is presented, together with the fit of the Fermi velocity, yielding 2.82 eV\AA ($v_F = 4.28 \times 10^5 \text{ m/s}$), a value similar to the one measured by Chen *et al.*³ (2.67 eV\AA , $v_F = 4.05 \times 10^5 \text{ m/s}$) for the $\overline{\Gamma\text{K}}$ direction.

Figure 3 shows further ARPES data for two selected thicknesses, measured with both He I and Kr photon sources. In Figs. 3(c) and 3(d) large amount of the additional spectral weight can be seen, which does not belong to the V-like dispersion of the Dirac cone. Between 10 and 8.8 QL thickness this spectral weight exhibits an upward shift of >100 meV, which is unexpected taking into account the absence of similar shifts in bulk bands in Figs. 2(a)–2(c). Therefore, the observed spectral weight might be related to the second dispersive branch of the surface state, as predicted by the theoretical calculations⁹ [see Fig. 2(g)]. Small perturbations in the dispersion in panels (a) and (b) can also be found, possibly indicating the same effect, with the difference between the spectra taken at two different photon energies possibly related to the matrix elements effects, which may be further enhanced in low-dimensional systems.

Our spectra for thicker films [16–11 QL, Figs. 2(a) and 2(b)] are very similar to the ones of Li *et al.*⁴ measured on 2–5 QL films. In their interpretation, the large CBM pocket is related to the thickness of the film, as they have observed a decreasing CBM spectral weight with increased thickness.

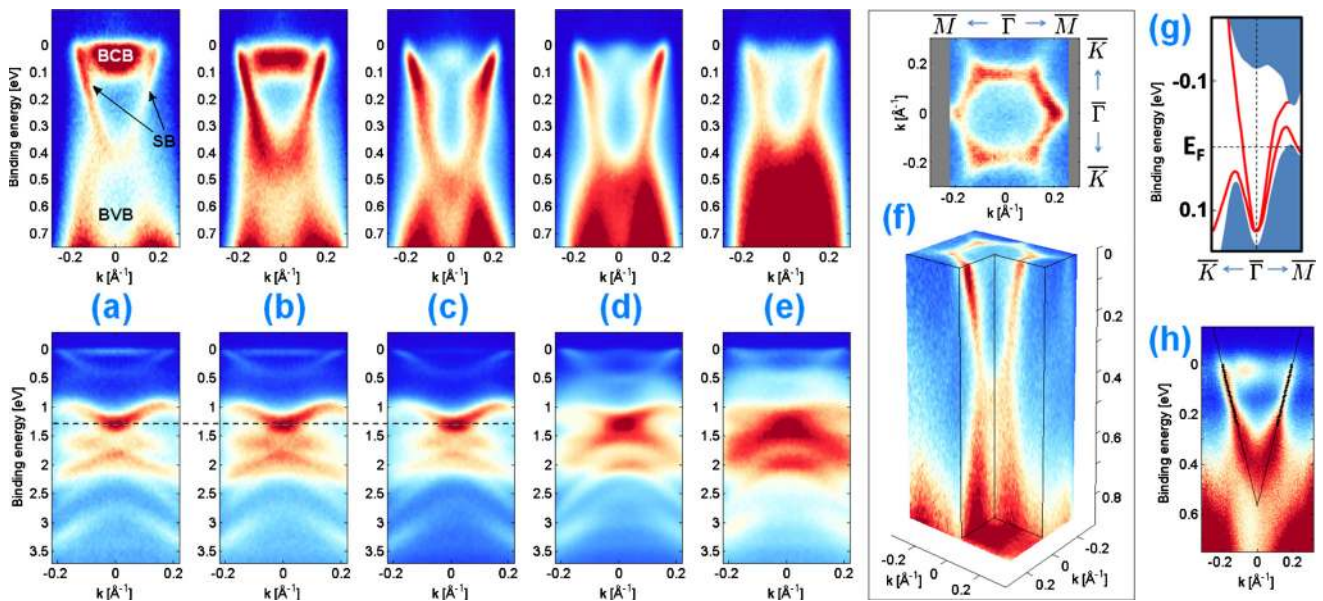


FIG. 2. (Color online) [(a)–(e)] Selected ARPES maps measured along the $\overline{\Gamma\text{K}}$ using He I radiation ($h\nu = 21.22 \text{ eV}$) at 140 K in the course of cleaning the same Bi_2Te_3 film several times. (a) 16 (≈ 16 nm), (b) 11, (c) 6, (d) 3.5, and (e) 1 QL. Bulk conduction band (BCB), surface state band (SB) and bulk valence band (BVB) are indicated in panel (a). Dashed line in bottom [(a)–(c)] panels indicate the unchanged binding energy position of the most pronounced bulk feature. The accuracy of thickness values is ± 0.5 QL (± 0.5 nm) to account for small instabilities of the sputter gun ion flux and a slightly nonuniform thickness profile. The color scale saturation is separately adjusted in the top and bottom panels. (f) Fermi surface and the three-dimensional band structure illustration of the 3.5 QL film measured at 15 K. (g) Schematic Bi_2Te_3 theoretical dispersion as predicted by Zhang *et al.*, Ref. 12. (h) 11 QL spectrum along $\overline{\Gamma\text{K}}$ taken with Kr (10.03 eV) excitation.

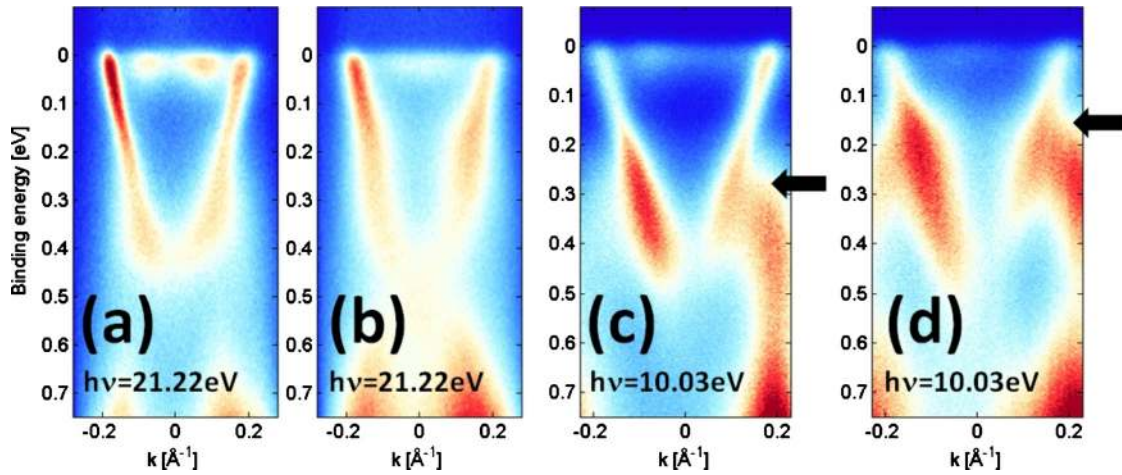


FIG. 3. (Color online) Bi_2Te_3 ARPES maps along $\overline{\Gamma K}$ measured at 90 K. (a) and (c) Film thickness is 10 QL, (b) and (d) Film thickness is 8.8 QL. [(a) and (b)] He I excitation, [(c) and (d)] Kr excitation. The >100 meV binding energy shift of the significant part of the spectral weight between panels (c) and (d) is indicated by arrows.

Since in our data exactly the opposite trend is observed, we postulate that the effect is related to the surface stoichiometry, which in Ref. 4 may be due to the time dependence of the film's growth, and in our case to the selective Ar-ion etching.

Experimental data presented in Figs. 2 and 3, are representative results obtained on different samples cut from the same wafer. After cleaning procedure all measured samples have shown Dirac cones of the same quality, and the CBM pocket could be removed in all cases.

In conclusion, we have shown that the surface of $\text{Bi}_2\text{Te}_3/\text{Si}(111)$ thin film can be effectively cleaned under UHV even after exposure to atmospheric conditions for many days. The cleaning process has been controlled by AES and recrystallization of the surface was confirmed by low energy electron diffraction (LEED). Photoemission spectra are of similar quality as those obtained on cleaved single crystals or on thin films never exposed to the air. Our results clearly indicate that the coherence of the Dirac-cone topological surface state in Bi_2Te_3 is, to a large extent, insensitive to the surface roughness induced by the sputter-and-anneal cleaning process, in agreement with other recent studies,^{5–8,15} which have shown insensitivity of the TI state to scattering on magnetic and nonmagnetic surface impurities. The observed trend of decreasing the CBM spectral weight with increasing sputtering time and decreased film thickness can be very promising from the point of view of further applications and indicates that the robust adjustment of the TI behavior might be achievable by a standard cleaning procedure under the UHV.

We acknowledge stimulating discussions with Caitlin Morgan on the course of editing the manuscript. Thanks are due to NRW Research School "Research with Synchrotron Radiation" funded by the Northrhine-Westphalia Ministry for Innovation, Science, Research, and Technology (Grant No. 321.2-8.03.06-58782).

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