Published in final edited form as:

Nano Lett. 2015 October 14; 15(10): 6365–6370. doi:10.1021/acs.nanolett.5b01358.

Robust Topological Interfaces and Charge Transfer in Epitaxial $\text{Bi}_2\text{Se}_3/\text{II-VI}$ Semiconductor Superlattices

Zhiyi Chen^{†,‡,*}, Lukas Zhao^{†,‡}, Kyungwha Park^{||}, Thor Axtmann Garcia^{‡,§}, Maria C. Tamargo^{‡,§}, and Lia Krusin-Elbaum^{†,‡}

[†]Department of Physics, The City College of New York, CUNY, New York, New York 10031, United States

[§]Department of Chemistry, The City College of New York, CUNY, New York, New York 10031, United States

[‡]The Graduate Center, CUNY, New York, New York 10016, United States

^{||}Department of Physics, Virginia Tech, Blacksburg, Virginia 24061, United States

Abstract

Access to charge transport through Dirac surface states in topological insulators (TIs) can be challenging due to their intermixing with bulk states or nontopological two-dimensional electron gas (2DEG) quantum well states caused by bending of electronic bands near the surface. The band bending arises via charge transfer from surface adatoms or interfaces and, therefore, the choice of layers abutting topological surfaces is critical. Here we report molecular beam epitaxial growth of $\text{Bi}_2\text{Se}_3/\text{Zn}_x\text{Cd}_{1-x}\text{Se}$ superlattices that hold only one topological surface channel per TI layer. The topological nature of conducting channels is supported by π -Berry phase evident from observed Shubnikov de Haas quantum oscillations and by the associated two-dimensional (2D) weak antilocalization quantum interference correction to magnetoresistance. Both density functional

*Corresponding Author: zchen2@ccny.cuny.edu.

The authors declare no competing financial interest.

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nano-lett.5b01358. Additional information and figures (PDF)

theory (DFT) calculations and transport measurements suggest that a single topological Dirac cone per TI layer can be realized by asymmetric interfaces: Se-terminated $\text{Zn}_x\text{Cd}_{1-x}\text{Se}$ interface with the TI remains “electronically intact”, while charge transfer occurs at the Zn-terminated interface. Our findings indicate that topological transport could be controlled by adjusting charge transfer from nontopological spacers in hybrid structures.

Keywords

Topological insulator; molecular beam epitaxy; superlattice; surface states; charge transfer; II–VI semiconductors

One of the main challenges in accessing surface states of three-dimensional (3D) topological insulators (TIs) is a significant conductivity in the bulk. Even in Bi_2Se_3 , which has the largest band gap (~ 0.3 eV)¹ among the family of second generation 3D TIs, electronic transport is dominated by charges donated by selenium vacancies and antisite defects, adding to thermal activation of charge carriers in the bulk. Bulk resistivity can be significantly increased using dopants such as Ca or Sb, but the downside is an additional sublattice disorder and possible clustering, both difficult to control.^{2,3} Electrostatic gating across the bulk bandgap in device structures based on TI thin films and crystalline flakes^{4–6} has been more promising, although surface oxidization and surface contamination can be a problem.^{7,8} One obvious way to decrease the contribution of the bulk is by increasing the surface-to-bulk ratio using nanostructuring which can be carried out through mechanical exfoliation, or growth of TI thin films, nanocrystals, or nanowires.^{9–11} While this approach has been also promising, transport measurements had to be made within 20 min postgrowth to minimize atmospheric doping^{10,12} and charge transfer from the ambient responsible for the so-called, surface “aging” effect.^{13,14}

In this work, we report on growth of $\text{Bi}_2\text{Se}_3/\text{Zn}_x\text{Cd}_{1-x}\text{Se}$ superlattices using molecular beam epitaxy (MBE). Since in a superlattice there will be multiple “surfaces”, the anticipated contribution from the surface states should be multiplied and easily accessed in charge transport or in optical measurements,^{15,16} if the topological properties of the interfaces are preserved. $\text{Zn}_x\text{Cd}_{1-x}\text{Se}$ was chosen to combine with Bi_2Se_3 with the expectation that chemical compatibility of the selenides will facilitate growth. In addition, it can be made lattice matched to Bi_2Se_3 , thereby reducing stresses, and has a wide bandgap and comparable growth conditions.¹⁷ The $\text{Zn}_x\text{Cd}_{1-x}\text{Se}$ layer could also work as a passivation layer, which would alleviate the problem of charge transfer from ambient adatoms. Here we demonstrate that Bi_2Se_3 layers and II–VI layers proximal to the TI layers are epitaxial single crystals and that the multilayered structure shows characteristics of topological surface transport. We also demonstrate that charge transfer from the Zn- but not Se-terminated II–VI layers ensures the asymmetry of “top” and “bottom” topological interfaces. This in turn guarantees that only one robust topological interface per TI layer contributes to 2D quantum transport and shows that, by choosing suitable spacer layers to modify charge transfer, topological surfaces in the hybrid structures can be controlled.

Superlattices were grown using two growth chambers connected by ultrahigh vacuum (UHV) transfer modules. The growth conditions we have developed for separate Bi_2Se_3

layers and $\text{Zn}_x\text{Cd}_{1-x}\text{Se}$ layers were reported previously.^{17,18} The steps in the growth protocol were as follows. First, a 10 nm Bi_2Se_3 layer was grown on a sapphire substrate, and the sample was transferred in vacuum to the II–VI chamber. Next, the growth of II–VI materials was performed under Se rich environment, with Se:group-II flux ratio of $\sim 10:1$. The ternary $\text{Zn}_x\text{Cd}_{1-x}\text{Se}$ was grown with the composition $x = 0.49$ that yields a near in-plane lattice match to Bi_2Se_3 .¹⁷ In what follows we will refer to this composition as ZnCdSe . The growth temperature was 270 °C, and the growth rate was ~ 7 nm/min. The sample was then transferred back to the Bi_2Se_3 growth chamber for the growth of next superlattice period.

Alternating Bi_2Se_3 and ZnCdSe layers were characterized in real time by reflection high-energy electron diffraction (RHEED). Figure 1a–d shows RHEED patterns observed during growth along one of the 6-fold symmetry axes of the Bi_2Se_3 surface. The sample shown here is a seven-period structure. The thicknesses of each ZnCdSe and Bi_2Se_3 layers are 12 and 10 nm, respectively. Figure 1a and c shows the patterns of the bottom-most and the top-most Bi_2Se_3 layer of the structure, while Figure 1b and d shows the ZnCdSe pattern along the same direction from the bottom-most and top-most ZnCdSe layers of the structure, as illustrated in Figure 1e. We conclude that there are no differences in the quality of the RHEED pattern between the bottom-most and top-most layers for both Bi_2Se_3 and ZnCdSe . In particular, the Bi_2Se_3 pattern appears very sharp and streaky in each repeat, suggesting that the surface recovers well each time Bi_2Se_3 is deposited. This indicates that the growth of samples with a large number of superlattice periods is feasible. The $(2\theta-\omega)$ high resolution X-ray diffraction (HR-XRD) scan shown in Figure 1f displays sharp peaks of $\text{Bi}_2\text{Se}_3(003)$, $\text{Bi}_2\text{Se}_3(006)$, and $\text{Bi}_2\text{Se}_3(012)$ as well as the sapphire(006) plane. The inset of Figure 1f shows a zoom-in view of the region of 2θ between 23° and 30° . From the zoom-in view, the large peak at $2\theta \sim 26.3^\circ$ can be assigned to the zero-order superlattice peak, $\text{SL}(0)$.¹⁹ A high-resolution transmission electron microscopy (HRTEM) image in Figure 1g demonstrates the remarkably sharp interfaces of a ZnCdSe layer sandwiched between two Bi_2Se_3 layers in a superlattice structure schematically shown in Figure 1h.

To investigate the effects of multilayering on charge transport, resistivity measurements were carried out on samples with Au electrical contacts lithographically patterned in a van der Pauw configuration shown in Figure 2a. The measurements were performed using Quantum Design Physical Property Measuring System (PPMS) in magnetic fields up to 14 T. In the low field regime ($B < 1$ T), the magnetoresistance of the superlattice sample decreases sharply as magnetic field decreases. The observed low-field cusp is a thumbprint of the weak antilocalization (WAL) quantum interference correction to “classical” resistivity, which in TIs is a signature of topological surface states originating from the topological π Berry phase.²⁰ Figure 2b shows sheet magnetoresistance for the seven-layer superlattice as a function of magnetic field component perpendicular to the ab -plane (normal to the (0001) surface), $B_\perp = B \cos \theta$. Here θ is the angle between field B and the c -axis of the Bi_2Se_3 layers. The data for all values of θ collapse on one curve this scaling of the WAL magneto-resistance cusp with $B_\perp$ demonstrates its 2D character.^{21,22} From the fit to Hikami–Larkin–Nagaoka (HLN) 2D localization theory²³

$$\Delta\sigma_{xx} = \sigma_{xx}(B) - \sigma_{xx}(0) \approx \alpha \frac{e^2}{2\pi^2\hbar} \left[\ln\left(\frac{B_\phi}{B}\right) - \Psi\left(\frac{1}{2} + \frac{B_\phi}{B}\right) \right] \quad (1)$$

where Ψ is the digamma function, we obtain the number of quantum 2D channels contributing to WAL. In theory, for a single 2D channel the coefficient α is $1/2$. B_ϕ in eq 1 is the dephasing magnetic field $B_\phi = \hbar/4el_\phi^2$ related to the dephasing length l_ϕ of interfering time-reversed electron paths. The fit yields $\alpha = 3.62 \pm 0.01$ (Figure 2c), which almost exactly corresponds to seven 2D conducting channels^{24,25} in our seven-layer superlattice. The result is robust and is reproduced in different superlattices (e.g., see Supporting Information Section B for data on a three-period superlattice). Notably, this is a first superlattice structure in which a single surface quantum channel per layer has been achieved.²⁶ From the fits to eq 1 we extract $B_\phi = 0.00705 \pm 0.00006$ T and calculate the dephasing length $l_\phi = 152.8 \pm 0.6$ nm (Figure 2c). l_ϕ decreases with increasing temperature (Figure 2d), following $l_\phi \propto T^{-0.46 \pm 0.03}$ in close correspondence with the $\sim T^{-0.5}$ dependence expected from theory in 2D systems.²⁷

To test the topological nature of the observed 2D magnetotransport we performed transport measurements at high fields applied normal to the superlattice's plane. Shubnikov de Haas (SdH) quantum oscillations in the longitudinal resistivity $R_{xx}(H)$ (R_{xx} is the longitudinal resistivity with the smooth background subtracted) are clearly observed (Figure 3a). By assigning Landau level (LL) index n to the minima in the oscillations,²⁸ we obtain the LL index plot shown in Figure 3b. The intercept β with the n axis in the high field limit gives a Berry phase of $2\pi\beta = \pi$, using $S_F(\hbar/eB) = 2\pi(n - \beta)$, where S_F is the extremal cross-section of the Fermi surface and factor $\beta = 1/2$. This is an indication of Dirac surface dominated transport.²⁹ The Fermi cross-section obtained from the period of SdH oscillations $S_F = 2.98 \times 10^{17} \text{ m}^{-2}$ corresponds to Fermi wavevector $k_f = 0.031 \text{ \AA}^{-1}$ and gives 2D carrier density of $7.56 \times 10^{11} \text{ cm}^{-2}$, which is comparable to the best results on single layers.³⁰ We note that the carrier mobility in our superlattices is an order of magnitude higher than reported previously in other superlattice structures;²⁶ we associate this with a “one-sided” interfacial charge transfer from ZnCdSe to Bi₂Se₃ as discussed below.

A single quantum channel per TI layer could indicate that top and bottom TI surfaces in each layer are coupled either through the conducting bulk⁹ or via coherent tunneling between them.^{25,31} Here, based on our density functional theory (DFT) calculations, we suggest another mechanism that could be at play. The idea is anchored on the inherent asymmetry of the interfaces of Bi₂Se₃ with wurtzite-structured¹⁷ ZnCdSe in the superlattices of interest. Geometries of wurtzite ZnCdSe and a superlattice are shown in Figure 1h. Inversion symmetry is absent in the wurtzite structure. When ZnCdSe is grown over the TI layer, it is likely that cation Zn or Cd is interfaced with the TI because of anionic nature of Se in the TI. Once a ZnCdSe layer is formed on the TI layer, the stoichiometry and charge neutrality of the ZnCdSe layer dictate Se termination just before another TI layer grows over the first ZnCdSe layer (Figure 1g).

DFT-calculated band structures for six quintuple-layers (6QLs) thick Bi₂Se₃ with Zn and Se terminated ZnSe layers and for bare 6QLs of Bi₂Se₃ are shown in Figure 4 (for details of the

method and the corresponding atomic structures see Supporting Information). The purpose of using $\text{Zn}_x\text{Cd}_{1-x}\text{Se}$ rather than ZnSe in the experiment was to reduce effects of stress caused by lattice mismatch at the interface. Since Cd also plays a role of cation like Zn, the replacement of $\text{Zn}_x\text{Cd}_{1-x}\text{Se}$ by ZnSe would not influence the charge transfer effect of our DFT calculations. The top and bottom TI surface states, colored as red and blue, respectively, in Figure 4a–c, are identified from electron charge density distribution as a function of vertical z coordinates.³² When the interface is terminated with cation Zn or Cd, as shown in Figure 4a, the Dirac cone at the interface substantially moves downward in energy relative to the bottom-surface Dirac cone,¹³ and the chemical potential (Fermi level) goes up and is shifted to the bulk conduction band. This is due to relaxation of interface geometry and charge transfer ($Q = 0.08e$ per 1×1 unit cell area) from the ZnSe layer to the topmost QL. The negative Q at the Zn-terminated interface (blue-hatched) in Figure 4f indicates charge transfer from Zn(1) dangling bonds to the topological surface. We remark that we have previously demonstrated that the wide band gap ZnSe or ZnCdSe overlayers can enforce quantum confinement of 2DEG quantum well states in Bi_2Se_3 .¹⁷ However, for the Se-terminated interface, as shown in Figure 4b, the Dirac cone at the interface does not change much relative the top-surface Dirac cone (only a slightly upward shift). The positive Q peak in Figure 4g suggests small charge transfer mostly in the interface region which is between Se(2) and Se(3) marked by the horizontal line. See Figures S4 and S5 for details of the relaxation of interface geometry and the charge transfer for both the Zn and Se-terminated structures. As a result of this interface asymmetry, we state that the top- and bottom-surface Dirac cones in the superlattice are well-separated. This separation between the two Dirac cones and the Fermi level crossing only the Dirac cone at the Se-terminated side is found in our superlattice calculation of the same thicknesses of the TI and ZnSe layers as the two slab calculations (for details see Supporting Information, Figure S6). Our proposed scenario would be consistent with a single 2D conduction channel per TI layer in the superlattice and could possibly account for the observed α values in other systems,^{9,25,31} although it should be noted that eq 1 is only an approximate description of weak localization and quantized bulk channels coupled to TI surfaces may also have to be considered.³³

The charge transfer is confirmed by our transport measurements on related structures. Three sets of samples were fabricated. The first set were two $\text{ZnSe}/\text{Bi}_2\text{Se}_3$ (10 nm)/sapphire layers, one of which was grown with Zn enriched interface (sample S1), while the other (sample S2) was grown under the standard protocol.¹⁷ In sample S1 bulk carrier concentration was found to be higher by ~55% and carrier mobility lower by ~29% than in sample S2 (Supporting Information, table in Section D). The second sample set comprised a thicker (250 nm) Bi_2Se_3 sample (S3) capped with ZnCdSe layer and a bare 250 nm Bi_2Se_3 layer (S4). Again, consistent with charge transfer from Zn termination apparent in the DFT calculations, we found that in S3 relative to S4 carrier concentration was increased by ~57%, and carrier mobility was reduced by ~20%.

To further test the effect of Zn at the interfaces, we have fabricated backgated structures using 200 μm thick SrTiO_3 (111) as a gate dielectric. Top-surface longitudinal resistivity R_{xx} vs gate voltage V_g measured at 1.9 K is shown in Figure 4h. Due to the large dielectric constant of SrTiO_3 at low temperatures, R_{xx} strongly depends on V_g .^{34,35} Figure 4h

demonstrates that a bare Bi_2Se_3 10 nm layer (sample S7) is clearly tuned by V_g across the Dirac point which corresponds to the maximum R_{xx} on the negative voltage side. This is similar to the experimental data reported previously.^{34,35} However, after an addition of Zn to the top surface (sample S6), ambipolar transport cannot be achieved since the Dirac point is not reached between -100 and 100 V in our experiments; i.e., the Dirac point is located far away from the Fermi level.

In conclusion, we have demonstrated a successful MBE growth of $\text{Bi}_2\text{Se}_3/\text{Zn}_x\text{Cd}_{1-x}\text{Se}$ superlattices. Crystalline II–VI band semiconductor layers with a degradation-resistant wurtzite structure¹⁷ were found to stabilize interfacial topological regions of Bi_2Se_3 layers. From Shubnikov-de Haas quantum oscillations we found that TI layers in a superlattice support a topological Dirac π -Berry phase.

In these TI/II–VI heterostructures we presented a first demonstration of a single two-dimensional quantum conduction channel per TI layer. Our DFT calculations and transport measurements provide strong evidence for asymmetric interfaces of II–VI layers with TI layers caused by charge transfer from the Zn-terminated top II–VI layers; this asymmetry is considered to be responsible for only one surface per layer contributing to 2D surface transport. One implication of our findings is that a growth process that could include Se irradiated interfaces may prevent charge transfer, thereby improving topological transport parameters. Thus, by using suitable nontopological electronically active spacer layers topological transport can be manipulated and complex nanodevices with vertical registry can be envisioned.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

This work was supported by the funds from NSF Grants DMR-1420634 (CCNY-Columbia MRSEC: Center for Precision Assembly of Superstratic and Superatomic Solids, PAS³), HRD-0833180 (CENSES) and DMR-1312483, and by DOD-W911NF-13-1-0159. K.P. was supported by NSF Grant DMR-1206354, computational support SDSC Trestles under DMR060009N, and Virginia Tech ARC.

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Figure 1.

RHEED patterns of the sample surface: a and c are of the bottom-most and the top-most Bi_2Se_3 layers. Panels b and d of the bottom-most and the top-most $\text{Zn}_x\text{Cd}_{1-x}\text{Se}$ layers, $x = 0.49$. The RHEED images indicate that there are no differences in the quality between the bottom-most layer and the top-most layer for both Bi_2Se_3 and $\text{Zn}_x\text{Cd}_{1-x}\text{Se}$. (e) A sketch linking RHEED patterns to their corresponding layers (yellow cylinder on the right represents gold contact across all layers for transport measurements). (f) $(2\theta-\omega)$ HR-XRD scan for the sample. Inset: expanded view of the SL(0) peak. (g) HRTEM image of a superlattice sample (grown on a sapphire substrate, S). Zn-terminated (red arrow) and Se-terminated (blue arrow) interfaces indicated in panel h are marked on the image. (h) Atomic structure of the superlattice, with wurtzite $\text{Zn}_x\text{Cd}_{1-x}\text{Se}$ abutted by Bi_2Se_3 (only 1QL is shown for simplicity). The horizontal axis is along the $[111]$ direction. The color scheme here is Zn (purple), Se (green), and Bi (pink). The two interfaces have different structural and electronic properties; see text.

Figure 2.

(a) A sketch of van der Pauw electrical contacts to all layers in the Bi₂Se₃ (gray)/ZnCdSe (blue) superlattice. Gold contacts at four corners and gold wires connected to them by silver paste are indicated. (b) Sheet magnetoresistance at 2K for the seven-layer superlattice plotted vs transverse component of magnetic field, $B_{\perp} = B \cos \theta$. Magnetoresistance (MR) scales with $B_{\perp}$ in the ± 1 T range, indicating the 2D character of the observed WAL induced by surface electrons and also a 2D character of the background “classical” MR, which is governed by mean free path (~ 10 – 20 nm, expectedly comparable to the TI layer thickness) rather than much longer (~ 150 nm) dephasing length l_ϕ . (c) Fit of the change in low-field conductance to 2D weak localization theory (eq 1). The shaded region highlights the low-field WAL range bounded by the dephasing field $B_\phi = 7.05$ mT $\ll 1$ T. (d) Dephasing length obtained from eq 1 vs temperature follows a power law (red) consistent with theory in 2D.

Figure 3.

High field SdH oscillations. (a) Traces of R_{xx} (blue) and derivative $dR_{xx}/d(\mu_0 H)$ (red) vs $1/B$ at $T = 1.9$ K at a magnetic field $H\parallel c$ up to 14 T (a smooth background has been subtracted). The magenta dashed lines indicate the minima of R_{xx} , and the green dashed lines mark the minima of $dR_{xx}/d(\mu_0 H)$. (b) Inverse magnetic field corresponding to the minima of R_{xx} (blue) and $dR_{xx}/d(\mu_0 H)$ (red) vs Landau level (LL) indices.

Figure 4.

DFT-calculated band structures for (a) 6QLs Bi₂Se₃ topped by a ZnCdSe layer with Zn at the interface, (b) 6QLs on top of a ZnCdSe layer with Se at the interface and (c) bare Bi₂Se₃ 6QLs. In a–c, the dashed lines indicate the Fermi levels E_F , and the top and bottom surface states are shown as red and blue, respectively. The arrows in a and b indicate bands strongly localized onto the top-most Se atoms in the ZnSe layer. (d) A sketch of the experimental structure, with different charge filling of Dirac cones. The top and middle layers in panel d are used for panel a, while the middle and bottom layers in panel d are for panel b. (e) Bare Bi₂Se₃ on sapphire has the same charge filling on both surfaces, as long as the substrate strain and surface adatom absorption are negligible. The blue Dirac cone in d and both Dirac cones in e have small fillings in the upper Dirac cones, unlike the DFT results, because pristine Bi₂Se₃ samples are typically *n*-type due to surface defects which were not taken into account in the DFT calculations. Calculated charge transfer Q between the TI layer and the (f) Zn-terminated and the (g) Se terminated ZnCdSe layer. Note that the vertical scale of panel f differs from that of g. Details of the charge transfer effect can be found in the Supporting Information. (h) Measured top-surface longitudinal resistivity R_{xx} vs gate voltage V_g for a Zn irradiated Bi₂Se₃ (sample S6, red dashed curve) layer and for a bare Bi₂Se₃ (sample S7, black solid curve) layer gated through SrTiO₃(111) at 1.9 K. The inset illustrates van der Pauw electrical top contacts.