Topological surface states usually emerge at the boundary between a topological and a conventional insulator. Their precise physical character and spatial localization depend on the complex interplay between the chemical, structural and electronic properties of the two insulators in contact. Using a lattice-matched heterointerface of single and double bilayers of β-antimonene and bismuth selenide, we perform a comprehensive experimental and theoretical study of the chiral surface states by means of microscopy and spectroscopic measurements complemented by first-principles calculations. We demonstrate that, although β-antimonene is a trivial insulator in its free-standing form, it inherits the unique symmetry-protected spin texture from the substrate via a proximity effect that induces outward migration of the topological state. This “topologization” of β-antimonene is found to be driven by the hybridization of the bands from either side of the interface.

Ever since the discovery of the importance of topology in condensed matter physics\(^1\), it has been demonstrated that the assembly of materials having different band structure topologies\(^2\) gives rise to a plethora of novel physical phenomena. The transformation of a topological insulator (TI) into a superconductor\(^3–5\), ferromagnetic TI\(^6\), or the appearance of Majorana fermions have been predicted\(^3\) and reported\(^7\). Topology has been invoked to explain the adsorbate/substrate resulting ferromagnetism\(^8–10\). Even graphene, in contact with a TI, shows effects related to spin–orbit coupling (SOC)\(^11,12\). These phenomena have been recognized as manifestations of a general “topological proximity effect”.

A conventional insulator (CI) and a TI, although sharing similar electronic structure in the bulk, at their interface feature a gap closing due to the different topological invariants\(^2\). This process typically occurs via the emergence of metallic topological surface states (TSS), when spatially moving from the CI to the TI. In this paper we have investigated the behavior of the TSS that generally arise at the interface between the two insulators. The localization of the TSS can undergo a shift from the TI to the first layers of the CI, it may stay put at the interface, or it can move back inside the TI\(^13\). In other words, if a TSS migration arises at the interface, it can be the result of a “topologization” of the CI or a “trivialization” of the TI\(^9,14–19\).

Gap size, SOC strength, work function difference, and relative thickness\(^20\) of the CI and TI layers contribute to determine the properties of the resulting interface, as they drive the hybridization among atomic orbitals at the two sides of the interface. Structural aspects are also important: huge changes in the electronic structure have been predicted\(^21,22\) and experimentally reported as being due to lattice strain and to the relative orientation of the two lattices\(^23\). A step forward has been made in the case of β-antimonene (i.e. the buckled honeycomb 2D allotrope of Sb) grown on bismuth selenide, where a perfect match at the interface is obtained with single orientation and negligible strain\(^24–26\). A single, buckled β-antimonene sheet will be hereafter referred to as a bilayer (BL).
The β-antimonene/Bi₂Se₃ hetero-structure constitutes an archetypal system for studying these phenomena. Experimental evidence of a TSS migration has only been explicitly reported at the Bi/TI interface, where the authors agree on the “topologization” of the bismuthene (i.e. honeycomb-like Bi) overlayer. However, one bilayer (BL) bismuthene hosts one-dimensional (1D) edge states typical of the quantum spin Hall phase (QSH), so that when deposited on top of a topological insulator, both 1D and 2D edge/surface states spatially coexist. In contrast, 1 BL and 2 BL of β-antimonene are trivial insulators while showing an important SOC as in the case of bismuth. This results in a simpler electronic structure, whose spin texture can be understood by atomistic calculations. This is of paramount importance in applications exploiting the electron spin, as tailoring the CI/TI spin textures can boost the realization of spintronics devices.

In the following we report on the atomic and electronic structures of atomically ordered β-antimonene layers on Bi₂Se₃(0001). These systems have been examined by scanning tunneling microscopy and spectroscopy (STM/STS), angle-resolved photoemission spectroscopy (ARPES) and ab initio calculations within the density functional theory (DFT). STM images reveal the formation of ordered single and double BL of β-antimonene on Bi₂Se₃, while STS of the unoccupied states reveal fingerprints of emergent topological bands. In both systems, ARPES measurements display well-defined dispersing bands, in good agreement with the calculated band structures. Hybridization between β-antimonene and Bi₂Se₃ determines the spin pattern of surface and interface electronic states. The formation of Dirac-cone surface-features demonstrates the topological character that β-antimonene inherits from the substrate via the proximity effect.

**Results and discussion**

The β-antimonene/Bi₂Se₃ hetero-structure constitutes an archetypal system for studying these phenomena. Experimental evidence of a TSS migration has only been explicitly reported at the Bi/TI interface, where the authors agree on the “topologization” of the bismuthene (i.e. honeycomb-like Bi) overlayer. However, one bilayer (BL) bismuthene hosts one-dimensional (1D) edge states typical of the quantum spin Hall phase (QSH), so that when deposited on top of a topological insulator, both 1D and 2D edge/surface states spatially coexist. In contrast, 1 BL and 2 BL of β-antimonene are trivial insulators while showing an important SOC as in the case of bismuth. This results in a simpler electronic structure, whose spin texture can be understood by atomistic calculations. This is of paramount importance in applications exploiting the electron spin, as tailoring the CI/TI spin textures can boost the realization of spintronics devices.

In the following we report on the atomic and electronic structures of atomically ordered β-antimonene layers on Bi₂Se₃(0001). These systems have been examined by scanning tunneling microscopy and spectroscopy (STM/STS), angle-resolved photoemission spectroscopy (ARPES) and ab initio calculations within the density functional theory (DFT). STM images reveal the formation of ordered single and double BL of β-antimonene on Bi₂Se₃, while STS of the unoccupied states reveal fingerprints of emergent topological bands. In both systems, ARPES measurements display well-defined dispersing bands, in good agreement with the calculated band structures. Hybridization between β-antimonene and Bi₂Se₃ determines the spin pattern of surface and interface electronic states. The formation of Dirac-cone surface-features demonstrates the topological character that β-antimonene inherits from the substrate via the proximity effect.
Figure 2. Computed band structures. (a) Bi$_2$Se$_3$, (b) 1 BL and (c) 2 BL of β-antimonene on Bi$_2$Se$_3$ along \( \bar{K} - \bar{\Gamma} - \bar{M} \) at the PBE-SOC-D2 geometry. The blue-to-red color gradient indicates the relative projection onto the BS front surface and Sb layers, respectively. Solid grey areas are the projected bulk bands. d Energy position at \( \bar{\Gamma} \) of TSS$_{\text{Sb}}$ (crosses) and P$_{\text{Sb}}$ (triangles) for the 1 BL case using slab geometries computed with different schemes. The grey columns indicate the projected bulk conduction band at \( \bar{\Gamma} \).

and lead to strong misalignments of β-antimonene and Bi$_2$Se$_3$ bands. Figure 1b also demonstrates the importance of accounting for SOC when computing the geometry. SOC reduces the inter-layer spacing \( d \) and has thus a similar effect as the vdW term. Since the semi-empirical D2 method yields results in good agreement with heavier \textit{ab-initio} vdW flavours, and correctly describes the bulk Bi$_2$Se$_3$ geometry, we adopt henceforth the PBE + SOC + D2 geometries.

Band structures of clean Bi$_2$Se$_3$, and with 1BL and 2BL of β-antimonene are reported in Fig. 2a–c along the \( \bar{K} - \bar{\Gamma} - \bar{M} \) symmetry lines. The color scale of the bands indicates their spatial origin by means of projection onto atomic orbitals of (red) β-antimonene or (blue) Bi$_2$Se$_3$ surface (defined here as half the 6 QL slab). This allows to distinguish truly surface-localized states of Bi$_2$Se$_3$ from bulk states (projected bulk in gray). Orange/yellow lines indicate hybridized states of mixed β-antimonene/Bi$_2$Se$_3$ (Sb/BS) character. The Fermi level (\( E_F \)) for both β-antimonene systems cuts the projected conduction band of bulk Bi$_2$Se$_3$, in contrast to the case of clean Bi$_2$Se$_3$, whose \( E_F \) is pinned at the Dirac point D of the topological surface state (TSS$_{\text{BS}}$). This reflects a charge transfer from Sb to the substrate upon deposition. Similar calculations have been reported previously for 1 BL and 2 BL of β-antimonene on Bi$_2$Se$_3$. We refer the reader to these works for a more detailed discussion of the origin of the band dispersion changes. Some important quantitative differences with respect to our calculations are discussed below.

Several bands are visible within the projected bulk gap. For the clean surface (Fig. 2a), the dominant feature is TSS$_{\text{BS}}$. The hetero-structures have more complicated band structures showing several hybridized bands for the single and double BLs. A topological surface state TSS$_{\text{Sb}}$ with corresponding Dirac point D* is evident in both systems. This state has a mixed orbital character for 1 BL and is of almost pure Sb-character near \( \bar{\Gamma} \) in the 2 BL system (in fact it is localized more strongly on the outermost Sb bilayer). The hybridization with Sb leads to modifications of the band dispersion around D*. For the 2 BL case, the effect of hybridization is so strong...
that the lower branch of the Dirac cone is unrecognizable. In both cases $D^*$ appears in the projected bulk local
gap above $E_F$, while the $TSS_{Sb}$ are clearly distinguished within the absolute gap, below $E_F$. A dispersing band $B$
of mixed orbital character is found below $TSS_{Sb}$ and merges with it at larger wave-vector ($k_{||}$) values for both Sb
coverages. A state of almost pure Sb character ($PS_{Sb}$) is found near the $Bi_2Se_3$ conduction band minimum for the
1 BL case, where also Rashba-split bands ($RBS$), mainly deriving from the substrate, appear beside and below
the bulk conduction bands. Similar Rashba bands are obtained for the 2 BL case, while $PS_{Sb}$ is shifted near the
valence band maximum.

Previous calculations of the 1 BL structure placed $D^*$ inside the $Bi_2Se_3$ conduction band and the apex of $P_{Sb}$
inside the bulk gap\textsuperscript{14}. We attribute this difference mostly to the omission of vdW interactions when optimizing
the hetero-structure geometry. Figure 2d indicates the energetic position of $P_{Sb}$ and $D^*$ at $\bar{\Gamma}$ for the 1 BL case,
as a function of the computational scheme adopted during the geometry optimization. The grey shaded area
indicates the projected bulk conduction band. In the case of pure PBE (no vdW treatment), both $D^*$ and $P_{Sb}$ lie
inside this band. Both points shift in energy, however, if vdW and/or SOC are included during the relaxation.
Fixing the substrate geometry to that of bulk $Bi_2Se_3$ causes a strong misalignment of these bands with respect to
the bulk bands (rightmost column). This stresses the importance of structural relaxation, dispersion and SOC
in studies of such topologically-hybridized hetero-structures. Further modifications of the band structures are
expected when quasi-particle corrections\textsuperscript{33–35} are considered.

Figure 3 shows (a) the STM image, (b) its DFT simulation, (c) the ball and stick model (top view, unit cell is
highlighted) and (d) the experimental and simulated STS spectra for 1 BL $\beta$-antimonene on $Bi_2Se_3$. The experimental
and simulated STM images are in very good agreement and clearly show a hexagonal pattern determined
by the upper Sb atoms ($Sb_2$ in Fig. 1a). An equivalent comparison for the 2 BL case is reported in Fig. 3e–h.
Although the buckled atomic structure of the topmost BL is almost identical to that of the 1 BL case\textsuperscript{26}, the STM
image is quite different, suggesting an important LDOS contribution to the STM images. The image is dominated
by bright spots associated with the topmost Sb atoms ($Sb_3$), but in this case a clear apparent buckled honeycomb
pattern is observed in both measured and computed STM images. Similar simulated images are obtained for different positions of $E_F$ from $-0.15 \text{ eV}$ to $+0.15 \text{ eV}$.

The simulated STM image in panel (f) reveals that the apparent buckling does not derive from the geometrical buckling of the outermost Sb BL. Indeed, the Sb atom (Sb$_1$) in the unit cell in Fig. 3g is imaged as a hollow in both the experimental and simulated STM images. From analysis of the integrated local density of states (LDOS), we find that TSS$_{Sb}$ strongly contributes to the 1 BL and 2 BL STM images reported in Fig. 3a,e (this is particularly true for the 1 BL case). The apparent buckling observed in the 2 BL STM images is actually determined by the shape and decay profile of the TSS$_{Sb}$ wave-function.

Figure 3d,h shows a comparison between the measured normalized differential conductance $dI/dV/(I/V)$ spectra for the β-antimonene layers and the computed LDOS. The grey shaded areas indicate the projected bulk bands at the $\Gamma$ point. The main experimental features are reproduced in the computed curves, such as the peaks at about $-0.32 \text{ eV}$ and $+0.18 \text{ eV}$ in the 1 BL case and the shoulders at $-0.02 \text{ eV}$ and $+0.38 \text{ eV}$ in the 2 BL spectra. Also evident is a "gap" in the 2 BL spectrum from about 0.05 to 0.23 eV that reflects a well-resolved minimum in the calculated data. The computed 1 BL curve in Fig. 3d shows a minimum very close to the position of the calculated $D^\ast$ ($0.23 \text{ eV}$): the alignment is not perfect due to the imposed broadening and nearby presence of the peak at 0.15 eV. Accordingly, the relative experimental curve shows a clear dip at the same energy, similarly to Sb on copper$^{36}$. The calculated 2 BL STS curve does not show a minimum in correspondence of the $D^\ast$ energy, since the LDOS is dominated by peaks of similar energy. A comparison with the computed band dispersions reveals that the four peaks and shoulders in the STS derive from key surface states of the β-antimonene/Bi$_2$Se$_3$ heterostructures. The peaks at $+0.18 \text{ eV}$ (1 BL) and $+0.38 \text{ eV}$ (2 BL) are fingerprints of the emergent topological surface states TSS$_{Sb}$, which exhibit strong curvature or even flattening in the vicinity of $D^\ast$. These features lie inside a gap in the projected bulk band structure as shown in Fig. 2b,c. In Ref. $^9$ a sharp peak in the STS spectrum of 2BL β-antimonene/Bi$_2$Se$_3$ at about 0.45 eV is attributed to a stationary point in the substrate conduction bands (their calculations place $D^\ast$ at $0.28 \text{ eV}$). However, their peak is well consistent with the sharp feature found at $+0.38 \text{ eV}$ in our computed LDOS arising from TSS$_{Sb}$ in the vicinity of $D^\ast$. This discrepancy may be due to the neglect of vdW interactions in Ref.$^24$. The shoulders at $-0.32 \text{ eV}$ (1 BL) and $-0.02 \text{ eV}$ (2 BL) derive from the bands which reverse slope at around 0.25 Å$^{-1}$ on either side of the $\Gamma$ point.

Figure 4 displays ARPES spectra of Bi$_2$Se$_3$ and β-antimonene/Bi$_2$Se$_3$ for different Sb coverages along the $\Gamma - K$ direction. Clean Bi$_2$Se$_3$ (Fig. 4a) shows a pair of linearly dispersing surface states near $E_F$ that are identified as TSS$_{Sb}$. D occurs at $-0.34 \text{ eV}$, in line with the results summarized in Ref.$^{37}$. Other intense bulk and surface features of Bi$_2$Se$_3$ are observed at larger binding energies. With the formation of 1 BL and 2 BL β-antimonene, new features appear in the Bi$_2$Se$_3$ band gap, while the substrate bands are attenuated and shifted to lower energies by the above discussed charge transfer process. Figure 4bc shows data corresponding to Sb coverages below (0.6 BL) and slightly above (1.1 BL) the completion of 1 BL, respectively. In both cases two dispersing bands are observed between $E_F$ and $-1 \text{ eV}$. They split upon approaching the zone center: one branch crosses $E_F$; the other reverses and slightly above (1.1 BL) the completion of 1 BL, respectively. In both cases two dispersing bands are observed in our computed LDOS arising from TSS$_{Sb}$ in the vicinity of $D^\ast$ (this is particularly true for the 1 BL case). The apparent buckling observed in the 2 BL STM images is actually determined by the shape and decay profile of the TSS$_{Sb}$ wave-function.

Figure 5 compares the experimental and calculated band structures of 1 BL and 2 BL on Bi$_2$Se$_3$ along $\Gamma - \bar{M}$ and $\bar{K} - \bar{\Gamma}$. In spite of the lack of quasi-particle corrections, and allowing for some misalignment of $E_F$, the overall agreement is rather good for 1 BL (Fig. 5a–d). The upper branch derives from the lower part of the TSS$_{Sb}$ band, while the lower branch can be identified with the band B. Both bands have a hybrid Sb/Bi$_2$Se$_3$ character. The Sb-related state $P_{Sb}$ with maximum at $-0.10 \text{ eV}$ at $\bar{\Gamma}$ cannot be clearly distinguished in the experiment, however. This can be attributed to the overlap with the substrate conduction band and to matrix element effects: according to DFT, the state has purely $p_x$, $p_y$ character and thus a low cross section in the photoemission experiments with p-polarized light in off-normal incidence.

Bands with an apparently similar dispersion in the proximity of $E_F$ were observed also for Sb layers grown on other TIs$^{38,39}$ and attributed to 1 Sb BL of unknown structure. However, the slopes of the bands for Sb layers on Sb$_2$Te$_3$ and Bi$_2$Te$_3$ are very different from the ones reported here and hardly compatible with the theoretical prediction. Theory predicts similar group velocities near $E_F$ for the TSS$_{Sb}$ bands on Sb$_2$Te$_3$ ($1.4 \text{ eV} \times \text{Å}$), Bi$_2$Te$_3$ ($1.7 \text{ eV} \times \text{Å}$) and Bi$_2$Se$_3$ ($1.3 \text{ eV} \times \text{Å}$) substrates$^{4}$. The group velocities measured for the TSS$_{Sb}$ bands in Ref.$^9$ were 3.1 $\text{eV} \times \text{Å}$ on Sb$_2$Te$_3$ and 2.5 $\text{eV} \times \text{Å}$ on Bi$_2$Te$_3$, i.e. almost twice larger than expected. In our DFT calculations we find a group velocity of approximately $1.5 \text{ eV} \times \text{Å}$, which is in perfect agreement with the one determined by the ARPES data in Fig. 5 ($1.6 \text{ eV} \times \text{Å}$).

Figure 5e–h shows the comparison of experimental and calculated band structure of 2 BL β-antimonene on Bi$_2$Se$_3$. The overall agreement is even better than for the 1 BL case. The larger splitting of the upper and lower branches along $\bar{K} - \bar{\Gamma} - \bar{K}$ than along $\bar{M} - \bar{\Gamma} - \bar{M}$ is well reproduced. Moreover, the $E_F$ crossing of these bands is also consistent. Only one band crosses $E_F$ along $\bar{K} - \bar{\Gamma} - \bar{K}$, while no crossing is seen along $\bar{M} - \bar{\Gamma} - \bar{M}$. From the orbital-projected band structure one can again identify the two strongly dispersive bands with the emergent TSS$_{Sb}$ band and the mixed origin of the B band. Furthermore, the band crossing near $-0.3 \text{ eV}$ at $\bar{\Gamma}$ (also present albeit less well resolved in the 1BL spectra) can be clearly interpreted as due to Rashba-split bands. According to the calculations, these bands are generated by the interface with β-antimonene but localized in the topmost
Bi$_2$Se$_3$ QLs. Their orbital character is very different from that of the original TSS$_{BS}$, being derived from the bulk conduction band. The Rashba splitting is caused by local dipole moments between QLs associated with the charge transfer from the $\beta$-antimonene to the substrate$^{40,41}$.

**Figure 4.** Experimental band structure. ARPES spectra measured with 20 eV photon energy along $\bar{K} - \bar{\Gamma} - \bar{K}$ of (a) Bi$_2$Se$_3$, (b) 0.6 BL, (c) 1.1 BL and (d) 2 BL of $\beta$-antimonene/Bi$_2$Se$_3$. 
We complete our analyses by considering the topological character of both systems as revealed by DFT. We verified that the free-standing 1 BL and 2 BL β-antimonene films are topologically trivial insulators by computing the $Z_2$ topological invariants (see Supplementary Information for details) following the parity method of Fu and Kane. This finding is consistent with previous calculations that predict a trivial-topological crossover to a quantum spin Hall phase for layers of at least 4 BLs. We also note that an odd number of bands cross the fundamental gap along $\Gamma - \bar{M}$, in both cases due to the single TSS branch that connects the bulk valence band to the bulk conduction band.

**Figure 5.** ARPES spectra and orbital-projected band structure. (a–d) 1 BL β-antimonene along the (first row) $\bar{M} - \Gamma - \bar{M}$ and (second row) $\bar{K} - \Gamma - \bar{K}$ directions. (e–h) Same as (a–d) for 2 BL β-antimonene.
The calculated spin texture of the clean Bi₂Se₃ and β-antimonene/Bi₂Se₃ hetero-structures is reported in Fig. 6. Figure 6b–f displays the $S_x$ and $S_y$ components of the spin expectation value along $\bar{K} - \bar{\Gamma} - \bar{M}$ (i.e. perpendicular to $k_x$ and $k_y$, as defined in Fig. 6a), projected on the β-antimonene layers (top row) and topmost Bi₂Se₃ QL (middle row), respectively. Zero polarization is found along the direction parallel to $k$ [See Fig. S3 and S4 of Supplementary Information]. Figure 6g–i shows the out-of-plane component of the spin expectation value $S_z$ along $\bar{K} - \bar{\Gamma} - \bar{K}$. While $S_z$ is practically zero in pure Bi₂Se₃, it becomes strong for the hetero-structures, even close to the $\bar{\Gamma}$ point for some bands. This may derive from a hexagonal warping effect induced by the β-antimonene layer or from considerable orbital momentum contributions. Several bands such as the band of mixed Sb/BS character labeled B, as well as TSSb in the 1 BL case, show $k$ dependent rotations in $S_y$. The B band in particular has a dominant $S_z$ component. In contrast, the Rashba-like character of RBS is confirmed by the spin texture which is completely confined to the substrate and lies fully within the surface plane (zero $S_z$ component). The second band crossing lying in the projected local bulk gap was clearly detected in the experiment (see around $-1.1$ eV in Fig. 4d) which likely corresponds to the Rashba-split bands seen here at $-0.8$ eV in the calculation. These latter bands have strong Sb character and thus have $S_z = 0$ only around the $\bar{\Gamma}$ point.
Spin analysis along the $\bar{K} - \bar{\Gamma} - \bar{K}$ direction (see Figs. S3 and S4 of Supplementary Information) confirms the helical nature of TSS$_{Sb}$. The 1 BL bands show a similar spin pattern to that of the substrate, wherein the spin magnetization density axis is locked perpendicular to $k$. For the 2 BL case, the spin polarization of TSS$_{Sb}$ within the surface plane shows the same spin-momentum locking, at least in a narrow range around $\bar{\Gamma}$ well above the $E_F$. The spin polarization of the TSS decreases from the center to the boundary of the Brillouin zone for the $S_x$ and $S_y$, while the opposite occurs for the $S_z$. This is generally ascribed to the superposition with bulk projected states.

TSS$_S$s thus displays the character of an emergent topologically protected state. By the comparison of the spin texture around the D (clean Bi$_2$Se$_3$) and D* points (either 1 BL or 2 BL), we note that the spin texture associated with the original TSS persists following hybridization with Sb states. In contrast, TSS$_{BS}$ is barely present in the Sb-covered surfaces (being hidden by the bulk valence band), implying that the state has lost its character (and topological protection) upon interfacing of the Sb layer.

The spin texture of TSS$_{Sb}$ derives mostly from the β-antimonene BL(s), although there is also a considerable contribution from the topmost QL in the 1 BL case, as expected from the orbital projection data (Fig. 2b,c). Figure 7a,b show TSS$_{BS}$ and TSS$_{Sb}$ in real space as isosurface plots of $|\Psi|^2$ at the $\bar{\Gamma}$ point and as plane-averaged plots, respectively. These plots at the D and D* points demonstrate the change in the character and localization of the topological state as a function of the Sb coverage. All three states have a similar total extension perpendicular to the surface (10–15 Å). The TSS$_{BS}$ state spans the whole topmost quintuple layer, as well as the first two atomic layers of the second QL, and derives from a mixture of Bi and Se orbitals. The TSS$_{Sb}$ is distributed across and outside the β-antimonene in the 1 BL system, with some contribution from the substrate (especially Se $p_z$ orbitals). Its depth profile is thus very similar to that of clean Bi$_2$Se$_3$, although it has clearly shifted towards the β-antimonene layer. The depth profile for 2 BL TSS$_{Sb}$ is quite different, being more strongly localized at the outermost β-antimonene BL and with weaker contributions from lower Sb and Se atoms. The different orbital origin of the two TSS$_{Sb}$ states conclusively explains the differences between their STM images in Fig. 3. The associated bands and spin texture around the D and two D* points (compare Fig. 6b,c,d) nonetheless remain strikingly similar.

These observations confirm that TSS$_{Sb}$ and the Dirac point at D* are truly emergent features and constitute a manifestation of a proximity effect in which an adsorbed trivial insulator (β-antimonene) inherits the unique electronic properties of a TI substrate (Bi$_2$Se$_3$). In other words, the helical state floats to the top of the CI (β-antimonene) which acquires a topological character. The hybridization with the substrate demonstrates that Bi$_2$Se$_3$ does not provide a platform for realizing free-standing β-antimonene, in spite of its vdW nature. However, it opens up the possibility to harness the topological nature of the Bi$_2$Se$_3$ substrate along with its momentum-locked spin character.

Conclusions
The preparation of large areas of single β-phase of antimonene on Bi$_2$Se$_3$ allowed to record unprecedentedly detailed ARPES and STM/STS reference spectra for 1BL and 2BL. Accurate quantum mechanical simulation including vdW and SOC have been performed to interpret the experimental data. The excellent agreement achieved with respect to the measured ARPES, STS, and STM data allows a consistent analysis of the experiments and provide a sound theoretical description of the underlying physical phenomena.

Our approach reveals a migration of TSS from the TI substrate to the outermost atomic layers of the CI. The migration occurs in a step-by-step manner as 1 and 2 BL of β-antimonene are adsorbed. In other words, the

Figure 7. The real space character of TSS$_{BS}$ and TSS$_{Sb}$. (a) Isosurfaces of TSS$_{BS}$ or TSS$_{Sb}$ at the $\bar{\Gamma}$ point and (b) their planar-averaged profiles.
particular electronic character of the β-antimonene/Bi₂Se₃ hetero-structures makes it possible to successfully extract the TSS from the TI surface and transfer it to the topmost atomic layers of the topologically trivial insulator. This behavior conforms the process of CI “topologization” in the hetero-structure, as distinct from the predicted “trivialization” of the TIs in MnSe/Bi₂Se₃, or the absence of migration of the TSS as predicted theoretically in ZnS/Bi₂Se₃. The spatially extended character of TSS in β-antimonene/Bi₂Se₃ demonstrates that it originates from the topology of bulk Bi₂Se₃, although orbital analysis reveals that it acquires considerable Sb character due to increasing hybridization. This origin appears to differ from previous suggestions.

The TSS migration is facilitated by the perfect lattice match of β-antimonene/Bi₂Se₃. Other vdW heterostructures with incommensurate interfaces, like a phase of antimonene on Bi₂Se₃, could hinder such migration due to the different interface potential, orbital hybridization, and local geometry, leading to topological states with different dispersion and localization. Lattice matching is therefore crucial in the design of vdW heterostructures with topological properties.

From a technological point of view, the β-antimonene/Bi₂Se₃ heterostructures considered here do not directly offer a route towards dissipationless currents, as bulk conduction bands open multiple scattering channels at Eₜ. It might be feasible to tune the position of the D⁺ points into the bulk fundamental gap by means of surface doping or external strain. Yet, the strong perturbation of the lower Dirac cones towards a Rashba-like character may offer alternative possibilities for probing electronic excitations of topologically protected helical states via inverse photoemission, two-photon photoemission, or optical techniques.

Finally, symmetry protection allows the migration of the TSS from a TI substrate to an adsorbate layer that is both elementally and structurally different. This includes the change in symmetry from the hexagonal Se layer to the honeycomb Sb, as well as the change from a passivated substrate to one with dangling bonds. This is relevant to all the cases where the surface of the TI is not suitable to an epilayer growth or to a functionalization. Ultimately, this may open a novel route towards easier handling of materials made of TI interfaced with metal contacts for the next generation of devices.

Methods
Sample preparation, STM/STS and ARPES measurements. Antimony was sublimated on in-situ cleaved Bi₂Se₃ at RT with a deposition rate of 0.5 Å/min, followed by a prolonged annealing at 473 K. The Sb coverage calibration was based on the known structural properties of the Ag/Sb surface alloy on Ag(111), which presents a (√3 × √3)R30° reconstructed surface. The structural ordering of the sample was confirmed by low-energy electron diffraction (LEED). STM images were measured by using an Omicron LT-STM instrument operating at 80 K. An electrochemically etched W wire was used as STM tip after electron bombardment cleaning in UHV. The STM scanner was calibrated measuring the clean Bi₂Se₃(0001) surface. The bias voltage is referred to the sample, hence positive (negative) bias corresponds to empty (filled) states. STS measurements have been acquired at 80 K using a lock-in amplifier with gap modulation at 5 kHz and 10 mV peak-to-peak amplitude. The STS spectra were collected on a 30 × 30 nm² image on a grid of 80 × 80 points. The reported spectra were obtained by averaging the normalized differential conductance curves (dI/dV)/(I/V) for 1 and 2 BL of β-antimonene. Photoemission experiments were performed at the VUV-Photoemission and BaDElPh beamlines of Elettra (Trieste, Italy). ARPES measurements were conducted at 80 K, with an angular resolution better than 0.3° and energy resolution of 30 meV.

Calculation method. DFT calculations were performed using the Vienna ab initio simulation package (VASP). Plane waves (kinetic energy cutoff of 400 eV) and projector augmented wave (PAW) pseudopotentials ([15 valence electrons for Bi (electronic configuration 5d¹⁰ 6s² 6p³), 6 valence electrons for Se (4s² 4p⁴), and 5 for Sb (5s² 5p³)]) were used, yielding well-converged (to 1 meV/Å) geometries and total energies. A Γ-centered 12 × 12 × 1 k-point mesh was used. The Bi₂Se₃ substrate was described using a 6 QL centrosymmetric slab, separated from periodically repeating replicas by thick (> 50 Å) vacuum regions. The experimentally determined atomic positions and lattice constants (a = 4.143 Å and c = 28.636 Å) were adopted. Sb atoms and the top four atomic layers of the substrate were relaxed. β-antimonene/Bi₂Se₃ hetero-structures were modelled by placing Sb atoms on both sides of the slab in their most stable geometries. The PBE exchange–correlation functional was employed with a correction to account for dispersion/vdW forces. The vdW approaches considered include Grimme’s semi-empirical schemes: D2, D3 without damping, and D3-BJ with Becke-Johnson damping; as well as the approach of Tkatchenko-Scheffler (TS) also with iterative Hirschfeld partitioning (TSH). Note that for bulk Bi₂Se₃, PBE-SOC-D2 yields interlayer distances very close to the experimental one. STM images were simulated via constant local density of states (LDOS) isosurfaces corresponding to the Tersoff-Hamann approximation. STS spectra were obtained by integrating the LDOS inside a 1.5 Å-thick box placed 2.5 Å above the topmost surface atom, and applying a broadening of 0.04 eV.

Data availability
The data that support the findings of this study are available from the corresponding authors upon request.

Received: 17 June 2020; Accepted: 19 August 2020
Published online: 03 September 2020

References
1. See for example: Asorey, M. Space, matter and topology. Nat. Phys. 12, 616–618 (2016).
2. Hasan, M. Z. & Kane, C. L. Colloquium: topological insulators. Rev. Mod. Phys. 82, 3045–3067 (2010).
5. Xu, J.-P.
8. Vobornik, I.
7. Nadj-Perge, S.
45. Gotlieb, K.
42. Fu, L. & Kane, C. L. Topological insulators with inversion symmetry. 
53. Sobota, J. A.
35. Förster, T., Krüger, P. & Rohlfing, M. Two-dimensional topological phases and electronic spectrum of Bi2Se3 thin films from GW calculations. 
28. Shoman, T.
38. Lei, T.
4. Maier, L.
3. Fu, L. & Kane, C. L. Superconducting proximity effect and majorana fermions at the surface of a topological insulator. 
50. Datzer, C.
46. Takayama, A., Sato, T., Souma, S. & Takahashi, T. Rashba effect in antimony and bismuth studied by spin-resolved ARPES. 
26. Hogan, C.
21. Huang, Z. Q.
22. Zhang, S., Yan, Z., Li, Y., Chen, Z. & Zeng, H. Atomically thin arsenene and antimonene: semimetal–semiconductor and indirect–direct band-gap transitions. 
24. Kim, S. H.
51. Reimann, J., Güdde, J., Kuroda, K., Chulkov, E. V. & Höfer, U. Spectroscopy and dynamics of unoccupied electronic states of the topological insulator Bi2Se3. 
23. Kim, S. H.
54. Panna, D.
14. Jin, K. H., Yeom, H. W. & Jhi, S. H. Band structure engineering of topological insulator heterojunctions. 
16. Menshchikova, T. V.
20. Costa, M.
21. Huang, Z. Q.
22. Zhang, S., Yan, Z., Li, Y., Chen, Z. & Zeng, H. Atomically thin arsenene and antimonene: semimetal–semiconductor and indirect–direct band-gap transitions. 
5. Xu, J.-P.
8. Vobornik, I.
7. Nadj-Perge, S.
45. Gotlieb, K.
42. Fu, L. & Kane, C. L. Topological insulators with inversion symmetry. 
53. Sobota, J. A.
35. Förster, T., Krüger, P. & Rohlfing, M. Two-dimensional topological phases and electronic spectrum of Bi2Se3 thin films from GW calculations. 
28. Shoman, T.
38. Lei, T.
4. Maier, L.
3. Fu, L. & Kane, C. L. Superconducting proximity effect and majorana fermions at the surface of a topological insulator. 
50. Datzer, C.
46. Takayama, A., Sato, T., Souma, S. & Takahashi, T. Rashba effect in antimony and bismuth studied by spin-resolved ARPES. 
26. Hogan, C.
21. Huang, Z. Q.
22. Zhang, S., Yan, Z., Li, Y., Chen, Z. & Zeng, H. Atomically thin arsenene and antimonene: semimetal–semiconductor and indirect–direct band-gap transitions. 
24. Kim, S. H.
51. Reimann, J., Güdde, J., Kuroda, K., Chulkov, E. V. & Höfer, U. Spectroscopy and dynamics of unoccupied electronic states of the topological insulator Bi2Se3. 
23. Kim, S. H.
54. Panna, D.
14. Jin, K. H., Yeom, H. W. & Jhi, S. H. Band structure engineering of topological insulator heterojunctions. 
16. Menshchikova, T. V.
20. Costa, M.
21. Huang, Z. Q.
22. Zhang, S., Yan, Z., Li, Y., Chen, Z. & Zeng, H. Atomically thin arsenene and antimonene: semimetal–semiconductor and indirect–direct band-gap transitions. 
24. Kim, S. H.
51. Reimann, J., Güdde, J., Kuroda, K., Chulkov, E. V. & Höfer, U. Spectroscopy and dynamics of unoccupied electronic states of the topological insulator Bi2Se3. 
23. Kim, S. H.
54. Panna, D.
14. Jin, K. H., Yeom, H. W. & Jhi, S. H. Band structure engineering of topological insulator heterojunctions. 
16. Menshchikova, T. V.
20. Costa, M.
21. Huang, Z. Q.
22. Zhang, S., Yan, Z., Li, Y., Chen, Z. & Zeng, H. Atomically thin arsenene and antimonene: semimetal–semiconductor and indirect–direct band-gap transitions. 
24. Kim, S. H.
51. Reimann, J., Güdde, J., Kuroda, K., Chulkov, E. V. & Höfer, U. Spectroscopy and dynamics of unoccupied electronic states of the topological insulator Bi2Se3. 
23. Kim, S. H.
55. Noakes, T. C., Hurt, D. A., McConville, C. F. & Woodruff, D. P. Structural investigation of ordered Sb adsorption phases on Ag(111) using coaxial impact collision ion scattering spectroscopy. Surf. Sci. 372, 117–131 (1997).
56. Petaccia, L. et al. BaDEIPh: A 4m normal-incidence monochromator beamline at Elettra. Nucl. Instrum. Methods Phys. Res. Sect. A 606, 780–784 (2009).
57. Kresse, G. & Furthmüller, J. Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set. Comput. Mater. Sci. 6, 15–50 (1996).
58. Kresse, G. & Furthmüller, J. Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. Phys. Rev. B 54, 11169 (1996).
59. Nakajima, S. The crystal structure of Bi2Te3-xSex. J. Phys. Chem. Solids 24, 479 (1963).
60. Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized gradient approximation made simple. Phys. Rev. Lett. 77, 3865–3868 (1996).
61. Grimme, S., Antony, J., Ehrlich, S. & Krieg, H. A consistent and accurate ab initio parametrization of density functional dispersion correction (DFT-D) for the 94 elements H-Pu. J. Chem. Phys. 132, 154104 (2010).
62. Grimme, S., Ehrlich, S. & Goerigk, L. Effect of the damping function in dispersion corrected density functional theory. J. Comput. Chem. 32, 1456–1465 (2011).
63. Tkatchenko, A. & Scheffler, M. Accurate molecular van der Waals interactions from ground-state electron density and free atom reference data. Phys. Rev. Lett. 102, 073005 (2009).
64. Bucko, T., Lebègue, S., Häfner, J. & Ángyán, J. G. Improved density dependent correction for the description of London dispersion forces. J. Chem. Theory. Comput. 9, 4293 (2013).

Acknowledgements
Calculations were performed at the Hazel Hen cluster at the Höchstleistungsrechenzentrum Stuttgart (HLRS), the Lichtenberg cluster at TU Darmstadt, the Center for Materials Research at JLU Gießen, and the CINECA Supercomputing Centre at Bologna. The authors acknowledge the computational resources and support provided by the HPC Core Facility and the HRZ of the Justus–Liebig Universität, and by CINECA via the ISCRA initiative. Technical support from Giovanni Emma, Massimiliano Rinaldi, Luca Sancin, and Fabio Zuccaro is acknowledged. We acknowledge the project EUROFEL-ROADMAP ESFRI. E.V.C. acknowledges support from the Saint Petersburg State University (Grant No. 51126254) and Fundamental Research Program of State Academies of Sciences for 2019–2021 (research direction III.23.2.9). The authors wish to thank Elettra Sincrotrone Trieste for providing access to its synchrotron radiation facilities.

Author contributions
S.K.M. and C.C. conceived the project. K.H. and C.H. performed the calculations and analysis with support from S.S. and E.V.C. S.K.M., P.M.S., P.M., R.F., A.B., M.P., and C.C. performed the ARPES measurements and analyses. F.R., S.C., and R.F. performed the S.T.M. and S.T.S. measurements and analysis. Z.S.A. and M.B.B. prepared the substrate. The manuscript was written by S.K.M., R.F., S.S., C.H., and C.C. and contributed to by all authors.

Funding
Open Access funding provided by Projekt DEAL.

Competing interests
The authors declare no competing interests.

Additional information

Correspondence and requests for materials should be addressed to S.K.M.

Reprints and permissions information is available at www.nature.com/reprints.

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2020