1. Introduction

Topology became a classification scheme for solid-state electronic properties in the 1980s while describing the robustness of the quantum Hall effect.\cite{1,2} This achievement has been honored most notably by the Noble prize 2016 for physics.\cite{3,4} The well-deserved appreciation was largely triggered by the experimental discovery of 2D topological insulators (2DTIs) in 2007.\cite{5} This discovery initiated a major effort in experimental and theoretical solid-state physics leading to a multitude of other types of topologies in crystalline solids, mostly appearing without magnetic fields.\cite{6–8} The overwhelming success has also led to activities in other fields of physics enabling, e.g., the guiding of light or sound along arbitrarily shaped edges.\cite{9–12} The attractive robustness of the topological properties, tied to the integer character of the topological indices, implied a multitude of proposals also for electronic applications.\cite{13–15} This currently culminates in the actively pursued dream to realize topological quantum computation via parafermions.\cite{16–19} The central advantage of this approach is the robustness of corresponding quantum operations against local perturbations as long as the quasiparticles remain in their topologically protected subspace.

From the materials science point of view, the intriguing observation that a lot of well-known materials are 3D strong topological insulators added a crucial view on electronic band structure properties.\cite{6–8} It turned out that a large amount of bulk insulators necessarily provide spin helical conductive surface states\cite{20–22} via the symmetry of their bulk band structure described by a topological index.\cite{23,24} The presence of such surface states is
totally independent on details of the confining surfaces and, moreover, these surface states are protected against backscattering by their spin helicity.\[6,25\] Hence, such materials can be thought of as a third conductivity class in addition to conductors and insulators, being insulating in the interior of the system but conducting on its surfaces. Favorably, a simple classification scheme exists in case of inversion symmetry of the crystal.\[25\]

It simply multiplies the parities (point inversion symmetries) of occupied single-electron states at the time-reversal invariant momenta (TRIMs) of the Brillouin zone to deduce the topological index. This provides an easy tool to exploit the much more complex theoretical background, that relies on extracting topological indices from general symmetries of the describing Hamiltonian\[23,24,26–28\] partly combined with crystal symmetries.\[29,30\] High-throughput density functional theory (DFT) calculations can be used to automatically extract candidate topological insulators from the extensive data base existing for crystalline materials.\[20,21\] This often leads to materials with large band gap such that the topological transport properties can be observed at room temperature.

However, subsequently, the candidate materials still have to be verified and characterized by experimental methods. This is due to the inherent minor difficulties of DFT calculations such as the missing precise description of electronic correlations and of van-der-Waals interactions\[31,32\] as well as the typically too small bandgap. As surface states are the decisive fingerprint of 3D topological insulators (3DTIs), well-established surface science methods became the tool of choice for the task of confirmation. In particular, angular-resolved photoelectron spectroscopy (ARPES)\[33,34\] directly maps the spin helical surface states in \( k \) space,\[33,34\] that typically exhibit a Dirac-type linear dispersion around one of its TRIMs in the Brillouin zone.\[35\] The spin-polarized version of ARPES (SARPES) moreover can characterize the spin-helical Dirac character of the topological surface states (TSSs).\[34\] Both can be compared directly with DFT-based calculations enabling an immediate verification of the topology.\[6–9\] Moreover, the doping level and, thus, the position of the intrinsic Fermi level \( E_F \) with respect to the Dirac point (DP) energy \( E_D \) can be checked via ARPES.\[34\] This is decisive for any type of applications in electronic devices requiring the Dirac cone to be present at \( E_F \).

For exploitations of topological insulators in electric transport experiments, it turned out that disorder is detrimental.\[17\] First, point defects acting either as acceptors or donors can make the interior conductive by shifting \( E_F \) into bulk bands.\[38,39\] Hence, the bulk conductivity often overwhelms the conductivity of the TSSs.\[40\] Second, surface doping can lead to a surface band bending that hosts additional 2D states of nontopological origin at \( E_F \), while the TSSs are detuned from \( E_F \).\[41,42\] The latter is difficult to avoid, as any contamination on the surface, resulting, e.g., from device preparation, can imply a band bending that even appears after a few minutes of ultraviolet (UV) illumination in ultrahigh vacuum (UHV).\[43\] Finally, even in case that \( E_F \) is favorably positioned within the bulk bandgap, compensation doping can lead to such strong potential fluctuations that electron and hole puddles appear in the interior of the sample implying hopping transport that competes with the transport via the TSSs.\[44,45\] Thus, experimental access to the potential disorder is crucial for improving the transport properties. The potential disorder can be mapped on small length scales by scanning tunneling spectroscopy (STS).\[46,47\] Therefore, one either uses the spatial variation of features in the local density of states (LDOS) related to \( E_F \) or the band edges\[48–50\] or, more precisely in energy, by spatially tracking Landau level energies in magnetic field.\[51–54\]

In addition, STS can map 1D topological states that are difficult to probe via ARPES,\[55,56\] as these states are typically only sparsely dispersed on the surface. STS identifies the topological edge states straightforwardly as the increased intensity of the LDOS at step edges.\[57–63\] Its distinctive property of prohibited backscattering appears via the missing standing waves. Such standing waves are very pronounced for conventional 1D electronic states due to the strongly confined 1D geometry.\[64\] Hence, its absence is a strong fingerprint of prohibited backscattering. 1D topological edge states have been found for 2D TIs,\[57,63,65\] weak topological insulators,\[58–60\] and at step edges of topological crystalline insulators, where they are caused by a symmetry breaking of the crystal at these edges.\[61\]

In this Short Review, we will exemplify the mentioned surface-science-based approaches to topology in crystals. These approaches are still central tools for the characterization of different topologies as well as for the finding of novel prospective materials within an established class of topology.\[66\] For the sake of simplicity, we restrict illustrations to our own work that cover many of the central developments yet. We cannot review the literature extensively, already due to the bare amount, but concentrate on initial publications and central additional insights on methodology.

2. Identifying Topological Surface States

Soon after establishing 2DTIs experimentally\[5\] based on theoretical predictions,\[26\] an extension of the formalism to 3D was proposed.\[24,25\] It results in two types of 3DTIs. One exhibits an odd number of spin-helical TSSs on each surface and is dubbed strong 3DTI, whereas the other one has an even number of TSSs on every surface except one and is dubbed weak 3DTI.\[24,25\] After identifying a first strong 3DTI in a BiSb alloy by ARPES,\[33\] DFT calculations predicted stoichiometric materials to be strong 3DTIs, namely \( \text{Bi}_2\text{Te}_3 \), \( \text{Sb}_2\text{Te}_3 \), and \( \text{Bi}_2\text{Se}_3 \).\[13\] These three materials share the same crystal structure of quintuple layers (QLs) that are stacked on each other by van-der-Waals forces.
Sb₂Te₃ is usually p-doped,[70,71] impeding ARPES mapping.

the TSS, but detrimental for electric transport. In contrast, ARPES measurements of a TSS on Bi₂Se₃(0001) have been published back to back with the DFT-based predictions.[35] First SARPES measurements appeared only 3 months later.[34] It turned out that the cleaved bulk samples of Bi₂Se₃ and Bi₂Te₃ are n-doped,[76] being beneficial for the ARPES mapping of the TSS, but detrimental for electric transport. In contrast, Sb₂Te₃ is usually p-doped,[70,71] imped ing ARPES mapping. Fortunately, we obtained a 20 year-old Sb₂Te₃ crystal that enabled establishing the favorable E₀ ≈ E_F. Similar results exhibiting Dirac cones within the bandgap close to E₀ have also been found for Sb₂Te₃, Bi₂Se₃, and Bi₂Te₃ after careful optimization of growth conditions in UHV.[69,73,75]

Figure 1e–g shows SARPES data recorded via a Mott detector, that probes the two in-plane directions of the spin. Two peaks at opposite k are recorded corresponding to the two opposite sides of the Dirac cone. The spin polarization is found to be exclusively perpendicular to the k vector as expected from the (Rashba-type) spin–orbit interaction. It is, moreover, helical, i.e., it switches sign when inverting k. These are the typical fingerprints of a Dirac cone type TSS.[35] Out-of-plane spin polarizations have also been observed, in particular, further away from E₀ and are traced back to distortions of the simple Dirac cone, e.g., via warping, i.e., by influences of the crystal structure.[76] It is important to realize that SARPES does not probe the spin polarization of the initial state, but that the photoemission process is an excitation to unoccupied states extending into the vacuum that can change the spin polarization either by matrix effects or by spin polarization of the final state.[77] This can be captured by calculations within the so-called fully relativistic one-step model based on
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In particular, at low photon energies, it turns out that the detected spin polarization can even be inverted with respect to the initial state depending on the polarization direction of the exciting light. At higher energies in the deep UV regime, this is less relevant, as excited states are well above the vacuum level. Hence, the helicity of the TSS can be deduced being counterclockwise for the lower part of the Dirac cone of \( \text{Sb}_2\text{Te}_3 \) (Figure 1d). This is in accordance with the DFT calculations (Figure 1b). The absolute value of the spin polarization of the TSS is not extracted directly from our SARPES data due to the limited angular and energy resolution. The reduced resolution during SARPES with respect to ARPES is caused by the low efficiency of the Mott detector. Novel approaches improve this efficiency considerably via spin dependent, \( \kappa \) conserving reflections of the photoelectrons at single crystals. Hence, resolution can be much better, but such apparatus was not available during the measurements shown in Figure 1. Consequently, spin polarization had to be extracted rather indirectly by carefully subtracting the inelastic background, the background originating from the also measured spin-polarized surface states at lower energy (visible in Figure 1b at \(-0.4 \text{ to } -0.8 \text{ eV}\)) and the background from the overlapping, enclosed bulk states. Nevertheless, the accordingly best fit of the SARPES data revealed a spin polarization of the TSS of \(80 - 95\%\) (Figure 1g) matching the DFT result of \(90\%\) surprisingly well. Obviously, the TSS is not \(100\%\) spin polarized, albeit it is spin helical. This is a natural consequence of spin-orbit interaction, that strongly mixes the spin with orbital degrees of freedom via the heavy atoms involved. Thus, spin is not a good quantum number in these materials.

3. Tuning the Dirac Point Energy

One main task after the experimental discovery of 3DTIs was to tune their DP energy \( E_D \), that mostly turned out to be far away from \( E_F \). Hence, literally speaking, the first 3DTIs were not even insulators in their interior. More importantly, the transport properties of the 3DTIs could not be probed without rendering the bulk of the material sufficiently insulating. A rather obvious, initial approach was to exploit the opposite p-type doping of \( \text{Sb}_2\text{Te}_3 \) and n-type doping of \( \text{Bi}_2\text{Te}_3 \) or \( \text{Bi}_2\text{Se}_3 \). Two main strategies have been pursued. Either, the two materials are mixed in a way such that they exhibit a similar density of acceptors and donors. This approach eventually led to the observation of the quantum Hall effect within thin films of \( \text{BiSbTeSe}_{2} \) as a clear signature of dominating 2D-type transport. A detailed analysis of the filling factor dependence of the Hall conductance identified the TSSs on bottom and top surfaces as the origin of the half integer quantum Hall effect. The respective tuning of the Dirac cone, namely of \( E_D \) with respect to \( E_F \), can be monitored by ARPES in detail. This is particularly important for the protection of Majorana states within vortices of a topological superconductor against conventional single-particle excitations by an effective gap \( E_{\text{gap,eff}} \) reading

\[
E_{\text{gap,eff}} \approx \dfrac{\Delta^2}{\sqrt{\Delta^2 + (E_F - E_D)^2}}
\]

with \( \Delta \) being the excitation gap of the surrounding topological superconductor.

Figure 2a shows the results for \( (\text{Bi}_{1-x}\text{Sb}_{x})_2\text{Te}_3 \) at different mixing of Bi and Sb including the case of \( E_F \approx E_D \). Corresponding ARPES data at \( x_{\text{Sb}} = 94\% \) are shown in Figure 2b. Figure 2c–f shows the evaluation of the data. The \( k \) values of the TSS are obtained from fitting intensity profiles \( I(k_i) \) (Figure 2c) at different energies that are subsequently extrapolated linearly to determine the crossing point as \( E_D \) (Figure 2d). Alternatively, the full width at half maximum (FWHM) of \( I(k_i) \) closer to \( E_D \) (Figure 2e) is used via identifying \( E_D \) as the energy with lowest FWHM (Figure 2f). In both cases, \( E_D \) has to be carefully calibrated as well. For the particular sample, we found \( E_D \approx E_D \) within \( 5 \text{ meV} \). As no time-dependent band shifts were observed, the value is likely robust as long as the sample is in UHV. However, ex situ Hall measurements on identically prepared samples exhibit a transition form p-type to n-type bulk conduction at much lower Sb concentration (\( x_{\text{Sb}} \approx 60\% \)). Hence, rescuing the precise tuning for electric devices requires additional efforts and investigations.

Another approach uses the electric field at interfaces between p-type and n-type 3DTIs. As well known for semiconductor p–n junctions, a depletion region forms at the interface such that a thin enough overlayer can maintain in the depletion region. This implies that \( E_F \) remains in the bandgap up to the surface. The approach has the general advantage that it avoids ternary or quaternary alloys that potentially induce additional scattering centers for electrons via alloying. Figure 3a shows a transmission electron microscope (TEM) image of a stack of n-type and p-type 3DTIs. Corresponding ARPES data at different thicknesses of the upper p-type \( \text{Sb}_2\text{Te}_3 \) on n-type \( \text{Bi}_2\text{Te}_3 \). Obviously, the Dirac cone is shifted downward in energy with decreasing \( \text{Sb}_2\text{Te}_3 \) thickness. To determine \( E_D \) including the thicknesses, it is above \( E_F \). DFT results of 6 QL \( \text{Sb}_2\text{Te}_3 \) are overlaid after rigidly shifting them to reproduce the ARPES data. It turned out that the best anchor point for shifting is the surface state at lower energy (Figure 1b at \(-0.4 \text{ to } -0.8 \text{ eV} \)). This state is vertically stronger confined to the surface area and, hence, is more intense in ARPES and less prone to the averaging by the vertical band bending details in the study by Eschbach et al. The resulting \( E_D - E_F \) has been compared with the result of a 1D Poisson–Schrödinger model revealing reasonable agreement (Figure 3c). The model is based on the charge carrier densities of MBE grown films of \( \text{Sb}_2\text{Te}_3 \) and \( \text{Bi}_2\text{Te}_3 \) as determined by Hall measurements, while assuming the same density of dopants and charge carriers. An intermixing at the interface is additionally considered that is deduced from Auger electron spectroscopy depth profiling. Obviously, the depletion method via p–n junction is also able to tune \( E_F \approx E_D \) for a thickness of \( \approx 20 \text{ QL} \) \( \text{Sb}_2\text{Te}_3 \) on top of \( \text{Bi}_2\text{Te}_3 \).

4. Materials with Particular Properties: Dual Topological Insulators and Phase Change Materials

Another important application of ARPES is to confirm desired properties of novel topological materials. This includes
topologically crystalline insulators (TCIs),[93,94] Dirac semimetals[95–97] and Weyl semimetals.[98,99] Interestingly, the topological properties of different kinds can be combined in a single material, if the topological indices belong to different symmetries of the Hamiltonian.[100] For example, 3DTIs protected by time-reversal symmetry can be combined with TCIs protected by a

![Image](https://www.advancedsciencenews.com/)

**Figure 2.** Tuning the DP by stoichiometry. a) Model of Dirac cone (blue, red lines) between the valence band (VB) and conduction band (CB) marked as gray areas for (Bi1−xSbx)2Te3 at different x. Resulting DP energy E_D with respect to E_F is indicated by the black dots. b) E(k) dispersion along K−Γ−K (dark: high intensity, bright: low intensity) for x_Sb = 94%. hν = 21.2 eV. Marked E_F is determined on polycrystalline Cu. c) Cut through (b) (pink dots) at E = E_F = −145 meV with fit curve (blue) consisting of two Voigt curves (black, red lines with indicated peak positions Max 1, Max 2). d) Energy-dependent peak positions (Max 1, Max 2) deduced from fits as shown exemplarily in (c) (red points with surrounding ellipses that enclose the 2σ confidence area). A linear regression of the data points (black, red dashed lines) is used to determine E_D = E_F = 2/67 meV. e) Cut through (b) at E = E_F (pink dots) exhibiting only a single peak. One Voigt fit (blue) is used to deduce FWHM. f) FWHMs of single peaks as in (e), i.e., close to E_F. Error bars are marked. T = 50 K. Adapted with permission.[89] Copyright 2015, AIP Publishing.

![Image](https://www.advancedsciencenews.com/)

**Figure 3.** Tuning the DP by band bending. a) High-angle annular dark-field (HAADF) cross-sectional image recorded by TEM of Bi2Te3/Sb2Te3 stack grown by MBE on Si(111). Different brightnesses indicate chemical contrast via different atomic weights of Bi and Sb. Inset: zoom with overlaid symbols representing Te atoms (red), Sb atoms (green), and Bi atoms (blue), T = 300 K. b) ARPES data (blue–white–red color code) of different numbers of Sb2Te3 QLs (marked above) on top of Bi2Te3/Si, hν = 8.4 eV, T = 15 K. The first two (last three) data sets are recorded along K−Γ−K (M−Γ−M). “Sb2Te3 only” marks thick Sb2Te3 on Si without Bi2Te3. DFT data of 6 QL Sb2Te3 (see also Figure 1b) are overlaid (symbols) after shifting in energy to optimally match experimental data. The resulting DP is marked. c) DP energies from ARPES (symbols) in comparison with a numerically solved 1D Poisson–Schrödinger model (full line). Inset: dopant densities used for the calculations as determined independently for both materials via Hall measurements (Bi2Te3: n-type N_D = 2 × 10^19 cm^-3, Sb2Te3: p-type N_A = 2 × 10^18 cm^-3). A linear interface intermixing across 5 nm is included as deduced from Auger electron spectroscopy depth profiling. Adapted under the terms of a Creative Commons Attribution 4.0 International License.[91] Copyright 2015, Springer Nature.
The bandgap of 0.1 eV around Bi1Te1. It consists of stacked Bi bilayers (BLs) and Bi2Te3 QLs in a ratio of 1:2 as evidenced by TEM (Figure 4a,b). Bi BLs are well known to be 2DTIs\(^{57,103,104}\) such that the stacking of such bilayers at sufficiently low interlayer interaction would result in a weak 3DTI. The so-called dark surface without TSS is simply the Bi BL surface, whereas the edge states of the BL lead to the TSSs at all other surfaces. The Bi2Te3 layers can be thought of as spacer layers between the Bi bilayers or as 2DTI layers themselves. Indeed, DFT calculations find a small bandgap of 0.1 eV around \(E_F\) for the intrinsic, i.e., undoped, Bi1Te1 with topological indices \((0;001)\). This indicates a weak 3DTI with its dark surface perpendicular to the \((001)\) direction.\(^{102}\) However, the reasoning via stacked 2DTIs is too simple, when analyzing the DFT data in more detail. Interlayer hybridizations mix up the 2D bands strongly, such that the weak 3DTI properties are rather accidental and not directly related to the 2DTI properties of the constituting layers. Intriguingly, the mirror Chern number of the same gap around \((1100)\) plane, is \(n_M = -2\) rendering the system a TCI as well. Consequently, one expects an additional pair of Dirac cones on the dark (001) surface of the weak 3DTI Bi1Te1. The DPs of these Dirac cones are necessarily located on the line in \(k\) space where the \((001)\) surface intersects with the \((1100)\) mirror plane. It must, moreover, be offset in opposite directions from \(\Gamma\). Figure 4c (upper row) shows a set of ARPES data in \(E(f_{k_{1,1}})\) representation that are recorded perpendicular to this mirror line for increasing \(f_{k_{1,2}}\) values along the line. The data exhibit an apparent Dirac cone as crossing of two bands at \(k_{1,2} \approx 0.18\ \text{Å}^{-1}\) and \(E - E_F \approx -0.2\ \text{eV}\). The development of the bands with \(k_{1,2}\) toward the crossing agrees nicely with corresponding DFT results of Bi1Te1(001) (Figure 4c, lower row). To achieve this agreement, the Bi1Te1 film had to be terminated by a single QL and had to be downshifted by 100 meV with respect to \(E_F\). Both is reasonable with the latter accounting for n-type doping as expected from the well-known n-type doping of Bi2Te3. The good agreement between ARPES and DFT data, also found for multiple other bands of Bi1Te1, is the central evidence for the dual topological character of Bi1Te1.\(^{102}\)

### 4.2. Topological Phase Change Materials

Another interesting class of 3DTI materials are commercially used in electronic applications. They are called phase change materials (PCMs) providing two favorable properties for data storage. First, they are fastly switchable (ns-scale) between the amorphous and a metastable crystalline phase at low energy penalty.\(^{105,106}\) Second, they exhibit a strong difference in optical reflectivity and electric conductivity between the two phases.\(^{107}\) Consequently, they are used in digital video disks (DVDs) and random-access memories.\(^{103}\) A standard class of PCMs is found on the pseudobinary line between the strong 3DTI Sb2Te3 and GeTe.\(^{108}\) The later material is also strongly influenced by spin–orbit interaction, revealing a strongly Rashba-split surface state and a strongly Rashba-split bulk state at \(E_F\).\(^{110-111}\) Hence, it is natural to assume that some of the PCMs are 3DTIs as well. Indeed, DFT predicts 3DTI properties\(^{112-113}\) and finds that the 3DTI character depends strongly on details of the atomic arrangement.\(^{114,115}\)

**Figure 4a** shows the structure of the most commonly used PCM Ge2Sb2Te5 in its metastable phase. It consists of alternating layers of hexagonal Te and a mixture of Ge, Sb and vacancies. Whether these layers, stacked in ABC-type fashion, exhibit additional order in the Ge/Sb/vacancy layers or not depends on details of the preparation and is decisive for the 3DTI properties.
according to DFT.\cite{112-115} Hence, a subtle borderline between strong 3DTI and trivial properties appears. Based on these findings, it has been speculated that the reversible, strong difference in electric conductivity of a superlattice GeTe/Sb$_2$Te$_3$, that appears after applying voltage pulses without making the material amorphous, is caused by a switch in topology.\cite{116,117}

However, the experimental confirmation of strong 3DTI properties in PCMs is complicated by the p-type doping. Hence, initially the only evidence by ARPES was the M-shaped valence band with maxima away from $\Gamma$ (Figure 5b).\cite{118} This configuration has been found in DFT calculations only for inverted bands of two Voigt peaks (smooth lines). Red, blue dots: peak positions of corresponding Voigt fits for several energies after averaging the cuts along two perpendicular $k$ directions. Red, blue lines: linear fits to the red and blue dots indicating a DP at $E_F$ - 160 meV. Adapted under the terms of a Creative Commons Attribution 4.0 International License.\cite{119} Copyright 2018, The Authors, published by Springer Nature.

5. Disorder Characterization

As described in Section 1, a central task for improving the electric transport properties of 3DTIs (and 2DTIs) is the reduction of disorder. Disorder can lead to additional transport channels concealing the features of the TSS as well as to scattering of the TSS electrons.\cite{8,37} STS is the tool of choice for probing the disorder at the surface due to its unprecedented spatial and energy resolution in probing the LDOS. It has only the minor drawback that it is exclusively measuring the surface disorder, but not the disorder within deeper layers of the bulk of the crystal.\cite{123}

One possibility by STS is to track characteristic features of the energy-dependent LDOS.\cite{48,49} One measures $dI/dV(V)$ curves with $I$ being the tunnel current and $V$ being the voltage applied between tip and sample. Mostly, such curves are measured by lock-in technique, i.e., the tip-surface distance is stabilized at voltage $V_{stab}$ and current $I_{stab}$. Afterward, the feedback loop is switched off, such that the tip-surface distance remains constant, while the voltage is changed linearly and overlapped with an oscillating voltage of amplitude $V_{mod}$ that enables the phase

\[ \Delta \text{delay} \text{ the initially unoccupied TSS and a second light pulse with time delay } \Delta \text{ extracts photoelectrons from the now-occupied TSS.} \]
sensitive detection of ∂I/∂V via a lock-in amplifier. In first order, the resulting ∂I/∂V represents the LDOS (E - E_F). This gives direct access, e.g., to spatial variations of the bandgap for a semiconductor or insulator.

Figure 6a shows the (111) surface of the strong 3DTI Ge_{2}Sb_{2}Te_{5} exhibiting Te as the top layer with hexagonal atomic structure. Several large triangular bright protrusions appear on top of the atomic lattice (Figure 6b–d). They have been identified as subsurface defects by comparison with DFT data. The lateral size of the triangle increases with the depth of the defect below the surface. The particular sample grown by MBE exhibits a defect density of ≈1.5 × 10^{12} cm^{-2}. This implies a potential disorder due to the positive charging of most of the defects, in particular, vacancies. The ∂I/∂V curves (Figure 6e) show a bandgap of about 0.5 eV with the valence band onset being close to E_F in agreement with optical absorption and ARPES data (Figure 5b), respectively. The bandgap onset is spatially varying. It is quantified via the peak position of the numerically determined d^{2}I/dV^{2} (V) curves leading to a nearly Gaussian distribution of the spatially varying valence band onset with σ width of 20 meV (Figure 6f). We compare this with a simple model calculation randomly distributing positive point charges with a density identical to the charge carrier density determined by Hall measurements (Figure 6g). This leads to potential fluctuations on the surface with the same σ width as in the experiment (Figure 6h). It implies that the Coulomb centers of the charged acceptors (vacancies) dominate disorder in this sample. Interestingly, the LDOS does not vanish within the bandgap (Figure 6e), indicating the presence of in-gap surface states in agreement with the two-photon ARPES revealing a TSS (Figure 5f).

Another possibility to map potential disorder is Landau level spectroscopy, however, requiring a magnetic field. It exploits the Dirac-type spin chirality of the TSS, implying a so-called zeroth Landau level (LL0) that is tied to E_F, i.e., averaged across some of the upper QLs. The lateral spatial resolution of the method is largely given by the magnetic length. Figure 6i shows STM data of in situ-cleaved Sb_{2}Te_{3}(0001) featuring few defects that have been identified previously by comparison with DFT calculations as Sb substitutional vacancies in the Sb layer and surface states in agreement with the two-photon ARPES revealing a TSS (Figure 5f).

Figure 6. Mapping the disorder potential. a) STM image of in situ-transferred Ge_{2}Sb_{2}Te_{5}(111) grown by MBE on Si(111). The hexagonal atomic structure of the top Te layer is visible with inset at larger magnification, V = -0.5 V, I = 100 pA, T = 300 K. b–d) STM images of characteristic triangular protrusions indicating subsurface defects, V = -0.5 V, I = 100 pA, T = 300 K. e) Scaled ∂I/∂V(∫) curves recorded at adjacent locations and displayed in different colors, V_{stab} = -300 mV, I_{stab} = 50 pA, V_{mod} = 5 mV, T = 9 K. Inset: zoom into the region of the valence band maximum (VBM). f) Histogram of VB onsets V_{VB} as deduced from the peak energies in d^{2}I/dV^{2}(V) curves (blue bars). A dashed Gaussian fit with marked σ width is added. g) Vertical cut through the simulated electrostatic potential V_{eff}(x, y, z) for randomly distributed bulk acceptors (red dots) at density N_A = 3 × 10^{24} m^{-3} as deduced from Hall measurements. h) Histogram of the potential values V_{eff}(x, y) at the surface resulting from multiple simulations as in (g) (blue bars). A dashed Gaussian fit with marked σ width is added. i) STM image of in situ-cleaved Sb_{2}Te_{3}(0001) exhibiting the hexagonally arranged top Te layer with clover-shaped defects likely SbT_{3} (bright) and subsurface Vac_{Sb} (dark), V = 0.4 V, I = 1 nA, T = 6 K.

j) d^{2}I/dV^{2}(V) at B = 6.7 T showing Landau levels of the TSS marked by level index n at the dashed lines that result from Lorentzian fits of the peaks, V_{stab} = 0.3 V, I_{stab} = 400 pA, V_{mod} = 4 mV, T = 6 K. E_0 is located at Landau level n = 0. Inset: d^{2}I/dV^{2}(V) at B = 0 T (same position) with E_0 marking the minimum of the curve, V_{stab} = 0.3 V, I_{stab} = 50 pA, V_{mod} = 4 mV, T = 6 K. k) d^{2}I/dV^{2}(V) at B = 0 - 7 T as marked recorded on a different sample area as in (j) and offset vertically, V_{stab} = 0.3 V, I_{stab} = 100 pA, V_{mod} = 2 mV, T = 6 K. The vertical dotted line indicates Landau level n = 0, hence, E_d. a–h) Ge_{2}Sb_{2}Te_{5}. Adapted with permission. Copyright 2017, American Physical Society. i–k) Sb_{2}Te_{3}. Adapted with permission.

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6. Edge States of Weak Topological Insulators

Weak 3DTIs have initially barely been studied due to the wrong conjecture that they are unstable with respect to most type of perturbations. More detailed studies, however, revealed that the only detrimental perturbation is a strong dimerization of adjacent layers along the surface normal of the dark surface leading to a doubling of the unit cell. Hence, also weak 3DTIs typically exhibit robust spin-helical surface states protected from backscattering. The most simple way to construct a weak 3DTI is stacking 2DTIs without interlayer interaction. This naturally implies that single-layer terraces on the dark surface are patches of 2DTIs that consequently must host 1D topological edge states at its step edges. These edge states are spin helical and, hence, ideal conductors as long as time-reversal symmetry is not broken. It turns out that such edge states appear generally for weak 3DTIs even if constructed differently. This implies the possibility to scratch a network of ideal conductors into the surface of a weak 3DTI.

The first experimental realization of a weak 3DTI was Bi₄Rh₃I₉. It consists of alternating layers of the 2DTI (Bi₄Rh)₃I₂⁺ and the trivial insulator Bi₂I₈ (Figure 7a). The 2DTI exhibits a honeycomb unit cell such as graphene, but is made of the heavy atoms Bi, I, and Rh (Figure 7b). It, thus, mimics the initial idea of a 2DTI in a honeycomb lattice, but provides a much stronger spin–orbit interaction leading to a sizable inverted bandgap of 200–300 meV. This gap is much larger than in graphene with inverted bandgap of ≈20 μeV. Hence, the idea to construct the 3D material is to stack 2DTI honeycomb structures that are separated by trivial insulators as spacers impeding interactions between the 2DTI layers. However, it turned out that the strong spin–orbit interaction shifts much more bands across $E_F$ than only the initial Dirac cone of the honeycomb lattice that appears at $E_F$ without spin–orbit interaction. Thus, the topological indices of a weak 3DTI again appear rather accidentally via inversion of several bands at the TRIMs of the Brillouin zone. Nevertheless, topological edge states at each step edge are expected and have been found by STS. They are directly visible as enhanced LDOS intensity at step edges (Figure 7c, background). In $dI/dV(V)$ curves, the bandgap region of the material (−0.15 to −0.35 eV) exhibits strong intensity exclusively at the step edges (Figure 7d). The edge states appear continuously along all edges and are only ≈1 nm wide perpendicular to the edge (Figure 7e). Moreover, the edge states did not exhibit any fingerprints of standing waves, but only intensity modulations periodic with the unit cell as expected for Bloch states. Thus, backscattering is largely impeded. Networks of topological edge states can indeed be scratched into the surface either by the tip of an atomic force microscope (AFM) with separation down to 25 nm or by the tip of an STM. The resulting scratches indeed show an increased LDOS within the bandgap (Figure 7g), but not at energies outside the gap (Figure 7h).

Unfortunately, $E_F$ is not within the bandgap and, thus, the edge states are not accessible by electric transport. Nevertheless, four-tip STM measurements in UHV have been applied. They revealed that the resistance as a function of distance between the tips is not described by a 3D transport model only, but required a sizable contribution from a parallel 2D transport channel (Figure 8b,c). The best fit of the experimental data (red curve in Figure 8c) implies conductances for the 2D and 3D contributions, $\sigma_{2D} = 0.064 \pm 0.005 S$ and $\sigma_{3D} = 920 \pm 800 S/m$, respectively. Thus, the 2D conductance corresponds to a ≈7 μm-thick layer with the 3D conductance $\sigma_{3D}$.

This implies that the surface region of Bi₄Rh₃I₉ is significantly more conductive than the bulk. The encouraging finding

![Figure 7](image-url)

**Figure 7.** Edge states on the dark side of a weak topological insulator. a) Atomic model of Bi₄Rh₃I₉ consisting of alternating layers of the 2D topological insulator (Bi₄Rh)₃I₂⁺ (red) and the trivial insulator Bi₂I₈ (blue). b) STM image recorded on a (Bi₄Rh)₃I₂⁺ terrace with atomic model structure overlaid using the same color code as in (b). V = 1.5 V, I = 100 pA. c) 3D perspective of adjacent STM image (front area, V = 0.8 V, I = 100 pA) and $dI/dV$ image (background, V = −0.337 V, I = 100 pA, $V_{mod} = 4$ mV), both recorded at the same (Bi₄Rh)₃I₂⁺ terrace confined by a step edge on the right. d) $dI/dV(V)$ recorded at a step edge of the (Bi₄Rh)₃I₂⁺ layer (grey), on a (Bi₄Rh)₃I₂⁺ terrace (red), and on a Bi₂I₈ terrace (blue), $V_{stab} = 0.8 V, I_{stab} = 100 pA, V_{mod} = 4$ mV. Notice the linearly vanishing $dI/dV$ intensity around $E_F$ caused by an Efros–Shklovskii-type Coulomb gap. (e) Stacked $dI/dV$ images recorded at the step edge of a (Bi₄Rh)₃I₂⁺ terrace for different V across the bandgap as marked on the left, $I_{stab} = 100 pA, V_{mod} = 4$ mV. f) Tapping-mode AFM image of Bi₄Rh₃I₉ after scratching a network of step edges into the surface by a carbon-coated Si cantilever at force $F = 1 \mu N$, $f_{res} = 275$ kHz, $k = 43$ N m⁻¹, $A = 30$ nm, set point: 70% (details see the study by Pauly et al. Copyright 2015, Springer Nature.)
is corroborated by DFT calculations of bulk Bi$_4$RhI$_9$ (Figure 8d, bottom, orange curve) showing $E_F$ within the bandgap. Additional calculations of a thin film revealed that the surface is strongly n-doped (Figure 8d, yellow curve) with the bandgap at similar energies as found in the STS data (Figure 8d, top, red curve).[143] This is in line with the strong 2D conductivity found by four-tip STM. The bandgap favorably moves quickly toward its bulk position already for the subsurface layer (Bi$_4$I$_3$). Bottom curve: layer resolved density of states of the 2DTI layer deduced from a DFT calculation (FPLO code[153]) of a slab with alternatingly stacked two 2DTI layers and two spacer layers (Bi$_4$I$_3$). Surface 2DTI layer is marked in pink for all four curves.[141] e) STM image of in situ-cleaved Bi$_4$RhI$_9$, $V = -0.1$ V, $I = 240$ pA, $300$ K. f) Scanning tunneling potentiometry image of the same area as (e) with indicated direction of applied current, $I = 1.2$ mA, distance of current carrying tips along vertical direction: $7.5$ μm, $300$ K. Part (d) adapted with permission.[141] Copyright 2016, American Chemical Society.

Consequently, the current induced potential is mapped. Figure 8e shows an area of the surface with 2DTI only, i.e., the islands exhibit step edges with height of a combined 2DTI and spacer layer. The potentiometry data show a barely visible overall decrease in the potential from the bottom to the top by about 0.1 mV due to the transport resistance. Much stronger features appear at the step edges and as patches on the surface of the 2DTI layers. They are identically present without applying current and are, hence, a static feature of the surface. Such features are caused by thermovoltage $V_{thermo}$ resulting from a temperature difference of tip and sample $\Delta T \approx 1$ K and, as such, indicate spatially different slopes of the LDOS at $E_F$ according to $V_{thermo} \propto \frac{\partial \text{LDOS}(E_F)}{\partial E} |_{E_F}$. As a result, the method reveals fluctuations of LDOS($E_F$) on the 2DTI terraces and a significant difference between step edges and terraces. The experiment did not provide any indication of preferred transport along the step edges in agreement with the observation that the topological edge states are not at $E_F$.

Other weak 3DTIs have been found,[99,60,147,148] but none with $E_F$ in the topological bandgap. Some uncertainty remains for ZrTe$_5$, that is very close to a topological phase transition such that details on strain and temperature change the topological properties partly also in a favorable way.[149] More interestingly, bismuthene, a honeycomb Bi monolayer on SiC(0001), is a 2DTI that can be prepared in UHV with $E_F$ inside the topological bandgap of size $\approx 1$ eV.[63] Here, preferential transport along step
edges might be detected by four-tip scanning tunneling potentiometry. Also, the ideal conductance of the edge state could be probed. It would lead to a potential drop that only appears at the end of the step edge, i.e., at the transition to the terrace in current direction.\cite{35}

7. Conclusions

In this article, we summarized some of the key contributions of surface science methods to the development of 3DTIs. Most importantly, ARPES could identify strong 3DTIs via the Dirac cone and its spin helicity of the TSS, whereas STS could identify weak 3DTIs via their helical edge states protected from backscattering at the dark surface. Moreover, ARPES was instrumental to monitor the tuning of the Dirac cone toward $E_F$, albeit the results are not compatible yet with the results from electric transport likely due to different treatments of the surfaces. Complementary, STS can map the potential disorder, most precisely via Landau level spectroscopy, and, hence, can monitor efforts to improve sample homogeneity. We have also shown exemplarily that particularly interesting materials can be identified as topological. In detail, we have discovered the first dual 3DTI Bi$_2$Te$_3$ and strong 3DTI properties in PCMs as an example material used in commercial applications. Two-photon ARPES was crucial to find the Dirac cone in these materials that only appeared in the unoccupied states due to strong p-doping. Finally, we have introduced the abilities of four-tip STM that can provide electric transport data in UHV without the requirement of ex situ contacting. We anticipate that this method will be perspective important to adapt the results from ARPES and STS to electric transport and, hence, to devices, as different surface treatments, that lead, e.g., to contaminations due to lithography, can be avoided.

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Conflict of Interest

The authors declare no conflict of interest.

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[1] D. J. Thouless, M. Kohmoto, M. P. Nightingale, M. den Nijs, Phys. Rev. Lett. 1982, 49, 405.
[2] Q. Niu, D. J. Thouless, Y. S. Wu, Phys. Rev. B 1985, 31, 3372.
[3] A. Cho, Science 2016, 354, 21.
[4] M. den Nijs, Science 2019, 364, 835.
[5] M. König, S. Wiedmann, C. Brune, A. Roth, H. Buhmann, L. W. Molenkamp, X. L. Qi, S. C. Zhang, Science 2007, 318, 766.
[6] M. Z. Hasan, C. L. Kane, Rev. Mod. Phys. 2010, 82, 3045.
[7] X. L. Qi, S. C. Zhang, Rev. Mod. Phys. 2011, 83, 1057.
[8] Y. Ando, J. Phys. Soc. Jpn. 2013, 82, 102001.
[9] B. Bahari, A. Ndao, F. Vallini, A. E. Amil, Y. Fainman, B. Kanté, Science 2017, 358, 636.
[10] J. Cha, K. W. Kim, C. Daraio, Nature 2018, 564, 229.
[11] Y. Yang, Z. Gao, H. Xue, L. Zhang, M. He, Z. Yang, R. Singh, Y. Chong, B. Zhang, H. Chen, Nature 2019, 565, 622.
[12] J. Ningyuan, C. Owens, A. Sommer, D. Schuster, J. Simon, Phys. Rev. X 2015, 5, 021031.
[13] X. Zhang, J. Wang, S. C. Zhang, Phys. Rev. B 2010, 82, 245107.
[14] L. He, X. Kou, K. L. Wang, Phys. Status Solidi RRL 2013, 7, 50.
[15] J. Han, A. Richardella, S. A. Siddiqui, J. Finley, N. Samarth, L. Liu, Phys. Rev. Lett. 2017, 119, 077702.
[16] J. Alicea, Rep. Prog. Phys. 2012, 75, 076501.
[17] S. D. Sarma, M. Freedman, C. Nayak, NPJ Quantum Inf. 2015, 1, 15001.
[18] J. Alicea, P. Fendley, Annu. Rev. Condens. Matter Phys. 2016, 7, 119.
[19] V. Lahtinen, J. Pachos, SciPost Phys. 2017, 3, 021.
[20] T. Zhang, Y. Jiang, Z. Song, H. Huang, Y. He, Z. Fang, H. Weng, C. Fang, Nature 2019, 566, 475.
[21] M. G. Vergniory, L. Elcoro, C. Felser, N. Regnault, B. A. Bernevig, Z. Wang, Nature 2019, 566, 480.
[22] F. Tang, H. C. Po, A. Vishwanath, X. Wan, Nature 2019, 566, 486.
[23] J. E. Moore, L. Balents, Phys. Rev. B 2007, 75, 121306.
[24] L. Fu, C. L. Kane, J. E. Mele, Phys. Rev. Lett. 2007, 98, 106803.
[25] L. Fu, C. L. Kane, Phys. Rev. B 2007, 76, 045302.
[26] C. L. Kane, J. E. Mele, Phys. Rev. Lett. 2005, 95, 146802.
[27] A. P. Schnyder, S. Ryu, A. Furusaki, A. W. W. Ludwig, Phys. Rev. B 2008, 78, 195125.
[28] S. Ryu, A. P. Schnyder, A. Furusaki, A. W. W. Ludwig, New J. Phys. 2009, 12, 065010.
[29] R. J. Slager, A. Mesaros, V. Juricic, J. Zanen, Nat. Phys. 2012, 9, 48.
[30] J. Kruthoff, J. de Boer, J. van Wezel, C. L. Kane, R. J. Slager, Phys. Rev. X 2017, 7, 041069.
[31] N. Mardirossian, M. Head-Gordon, Mol. Phys. 2017, 115, 2315.
[32] M. Stöhr, M. T. Vorhis, A. Tkatchenko, Chem. Soc. Rev. 2019, 48, 4118.
[33] D. Hsieh, D. Qian, L. Wray, Y. Xia, Y. S. Hor, R. J. Cava, M. Z. Hasan, Nature 2008, 452, 970.
[34] D. Hsieh, Y. Xia, L. Wray, D. Qian, A. Pal, J. H. Dil, J. Osterwalder, F. Meier, G. Bihlmayer, C. L. Kane, Y. S. Hor, R. J. Cava, M. Z. Hasan, Science 2009, 323, 919.
[35] H. Zhang, C. X. Liu, X. L. Qi, X. Dai, Z. Fang, S. C. Zhang, Nat. Phys. 2009, 5, 438.
[36] D. Hsieh, Y. Xia, D. Qian, L. Wray, J. H. Dil, F. Meier, J. Osterwalder, L. Patthey, J. G. Checkelsky, N. P. Ong, A. V. Fedorov, H. Lin,
A. Bansil, D. Grauer, Y. S. Hor, R. J. Cava, M. Z. Hasan, Nature 2009, 460, 1101.

[37] D. Culcer, Physica E 2012, 44, 860.

[38] B. Yan, D. Zhang, C. Felser, Phys. Status Solidi RRL 2012, 7, 148.

[39] P. Cheng, C. Song, T. Zhang, Y. Zhang, Y. Wang, J. F. Jia, J. Wang, Y. Wang, B. F. Zhu, X. Chen, X. Ma, K. He, L. Wang, X. Dai, Z. Fang, X. Xie, X. L. Qi, C. C. Liu, S. C. Zhang, Q. K. Xue, Phys. Rev. Lett. 2010, 105, 076801.

[40] T. R. Devidas, E. P. Amaldass, S. Sharma, R. Rajaraman, D. Sornadurai, N. Subramanian, A. Mani, C. S. Sundar, A. Bharathi, Europhys. Lett. 2014, 108, 67008.

[41] M. Bianchi, D. Guan, S. Bao, J. M. B. Iversen, P. D. King, P. Hofmann, Nat. Commun. 2010, 1, 128.

[42] P. D. C. King, R. C. Hatch, M. Bianchi, R. Ovsyannikov, C. Lupulescu, G. Landolt, B. Slomski, J. H. Dil, D. Guan, J. L. Mi, E. D. L. Rienks, J. Fink, A. Lindblad, S. Svensson, S. Bao, G. Balakrishnan, B. B. Iversen, J. Osterwalder, W. Eberhardt, F. Baumberger, P. Hofmann, Phys. Rev. Lett. 2011, 107, 096802.

[43] E. Frantzeskakis, S. Ramankutty, N. de Jong, M. Morgenstern, Rev. Mod. Phys. 2016, 88, 021004.

[44] B. Lampert, K. Reichelt, J. Cryst. Growth 1981, 51, 203.

[45] D. Teweldebrhan, V. Goyal, A. A. Balandin, Nano Lett. 2010, 10, 1209.

[46] Y. Xia, Q. Dian, D. Hsieh, L. Wray, A. Pal, H. Lin, A. Bansil, D. Grauer, Y. S. Hor, R. J. Cava, M. Z. Hasan, Nat. Phys. 2009, 5, 398.

[47] D. Hsieh, Y. Xia, D. Qian, L. Wray, F. Meier, J. H. Dil, J. Osterwalder, L. Patthey, A. V. Fedorov, H. Lin, A. Bansil, D. Grauer, Y. S. Hor, R. J. Cava, M. Z. Hasan, Phys. Rev. Lett. 2009, 103, 146401.

[48] C. Seibel, H. Maas, H. Bentmann, J. Braun, K. Sakamoto, M. Arita, K. Shimada, J. Minár, H. Ebert, F. Reintert, J. Electr. Spectrosc. Relat. Phenom. 2015, 201, 110.

[49] C. Pauly, G. Bihlmayer, M. Liebmann, M. Grob, A. Georgi, D. Subramanian, M. R. Scholz, J. Sánchez-Barriga, A. Varykhalov, S. Blügel, O. Rader, M. Morgenstern, Phys. Rev. B 2012, 86, 235106.

[50] G. Wang, X. Zhu, J. Wen, X. Chen, K. He, L. Wang, X. Ma, Y. Liu, X. Dai, Z. Fang, J. Jia, Q. Xue, Nano Res. 2010, 3, 874.

[51] Y. Jiang, Y. Y. Sun, M. Chen, Y. Wang, Z. Li, C. Song, K. He, L. Wang, X. Chen, Q. K. Xue, M. A. S. Bhang, Phys. Rev. Lett. 2012, 108, 066809.

[52] K. Hoefner, C. Becker, D. Rata, J. Swanson, P. Thalmeier, L. H. Tjeng, Phys. Rev. B 2016, 93, 104401.

[53] S. Komura, S. Souka, T. Sato, M. Komatsu, A. Takayama, T. Takahashi, M. Kriener, K. Segawa, Y. Ando, Phys. Rev. Lett. 2011, 106, 216803.

[54] S. Hüfner, Photoelectron Spectroscopy, Springer, Berlin Heidelberg 1995.

[55] H. Ebert, D. Kögderitzsch, J. Minár, Rep. Prog. Phys. 2011, 74, 096501.

[56] C. Jozwiak, C. H. Park, K. Gottlieb, C. Hwang, D. H. Lee, S. G. Louie, J. D. Denlinger, C. R. Rotundu, R. J. Birgeneau, Z. Hussain, A. Lanzara, Nat. Phys. 2013, 9, 293.

[57] Kitij Medjanik, O. Fedchenko, S. Chernov, D. Kutnyakov, M. Ellguth, A. Oelsner, B. Schönheine, T. R. F. Peixoto, P. Lutz, C. H. Min, F. Reintert, S. Däster, Y. Acremen, J. Viethaus, W. Huth, J. H. Elmers, G. Schönheine, Nat. Mater. 2017, 16, 615.

[58] Z. Ren, A. A. Taskin, S. Sasaki, K. Segawa, Y. Ando, Phys. Rev. B 2010, 82, 241306.

[59] D. Kong, Y. Chen, J. J. Cha, Q. Zhang, J. C. Analytis, K. Lai, Z. Liu, S. S. Hong, K. J. Koski, S. K. Mo, Z. Hussain, I. R. Fisher, Z. X. Shen, Y. Cui, Nano Lett. 2011, 11, 4693.

[60] Y. Xu, I. Miotkowski, C. Liu, J. Tian, H. Nam, N. Aldoust, J. Hu, C. K. Shih, M. Z. Hasan, Y. P. Chen, Nat. Phys. 2014, 10, 956.

[61] Y. Xu, I. Miotkowski, Y. P. Chen, Nat. Commun. 2016, 7, 11434.

[62] Y. Zhang, C. Zhang, Z. Zhang, J. Wen, X. Feng, K. Li, M. Liu, K. He, L. Wang, X. Chen, Q. K. Xue, M. Yang, Nat. Commun. 2011, 2, 574.

[63] J. D. Sau, R. M. Lutchyn, S. Tewari, S. D. Sarma, Phys. Rev. B. 2011, 83, 075422.

[64] A. L. Rakhmanov, A. V. Rozhkov, F. Nori, Phys. Rev. B 2011, 84, 051304.

[65] P. J. Xu, C. Liu, M. X. Wang, J. Ge, Z. L. Liu, X. Yang, Y. Chen, Y. Liu, Z. A. Xu, C. L. Gao, D. Qian, F. C. Zhang, J. F. Jia, Phys. Rev. Lett. 2012, 108, 217001.
[138] B. Voigtländer, V. Cherepanov, S. Korte, A. Leis, D. Cuma, S. Just, F. Lüpke, Rev. Sci. Instrum. 2018, 89, 101101.
[139] S. Just, M. Blab, S. Korte, V. Cherepanov, H. Soltner, B. Voigtländer, Phys. Rev. Lett. 2015, 115, 066801.
[140] S. Just, H. Soltner, S. Korte, V. Cherepanov, B. Voigtländer, Phys. Rev. B 2017, 95, 075310.
[141] C. Pauly, B. Rasche, K. Koepfermik, M. Richter, S. Borisenko, M. Liebmann, M. Ruck, J. van den Brink, M. Morgenstern, ACS Nano 2016, 10, 3995.
[142] M. P. Ghimire, M. Richter, Nano Lett. 2017, 17, 6303.
[143] P. Muralt, D. W. Pohl, Appl. Phys. Lett. 1986, 48, 514.
[144] F. Lüpke, S. Korte, V. Cherepanov, B. Voigtländer, Rev. Sci. Instrum. 2015, 86, 123701.
[145] J. A. Stovneng, P. Lipavský, Phys. Rev. B 1990, 42, 9214.
[146] T. Druga, M. Wenderoth, J. Homoth, M. A. Schneider, R. G. Ulbrich, Rev. Sci. Instrum. 2010, 81, 083704.
[147] M. M. Hosen, K. Dimitri, A. K. Nandy, A. Aperis, R. Sankar, G. Dhakal, P. Maldonado, F. Kabir, C. Sims, F. Chou, D. Kaczorowski, T. Durakiewicz, P. M. Oppeneer, M. Neupane, Nat. Commun. 2018, 9, 3002.
[148] R. Noguchi, T. Takahashi, K. Kuroda, M. Ochi, T. Shirasawa, M. Sakano, C. Bareille, M. Nakayama, M. D. Watson, K. Yaji, A. Harasawa, H. Iwasawa, P. Dudin, T. K. Kim, M. Hoesch, V. Kandyba, A. Giampietri, A. Barinov, S. Shin, R. Arita, T. Sasagawa, T. Kondo, Nature 2019, 566, 518.
[149] Y. Zhang, C. Wang, L. Yu, G. Liu, A. Liang, J. Huang, S. Nie, X. Sun, Y. Zhang, B. Shen, J. Liu, H. Weng, L. Zhao, G. Chen, X. Jia, C. Hu, Y. Ding, W. Zhao, Q. Gao, C. Li, S. He, L. Zhao, F. Zhang, S. Zhang, F. Yang, Z. Wang, Q. Peng, X. Dai, Z. Fang, Z. Xu, et al., Nat. Commun. 2017, 8, 15512.
[150] U. Klass, W. Dietsche, K. von Klitzing, K. Ploog, Surf. Sci. 1992, 263, 97.
[151] A. L. Efros, B. I. Shklovskii, J. Phys. C: Solid State Phys. 1975, 8, L49.
[152] F. Lüpke, S. Just, M. Eschbach, T. Heider, E. Mlyntczak, M. Lanius, P. Schüffelgen, D. Rosenbach, N. von den Driesch, V. Cherepanov, G. Mussler, L. Plucinski, D. Grützmacher, C. M. Schneider, F. S. Tautz, B. Voigtländer, NPJ Quantum Mater. 2018, 3, 46.
[153] K. Koepfermik, H. Eschrig, Phys. Rev. B 1999, 59, 1743.