In situ disentangling surface state transport channels of a topological insulator thin film by gating

In the thin film limit, the surface state of a three-dimensional topological insulator gives rise to two parallel conduction channels at the top and bottom surface of the film, which are difficult to disentangle in transport experiments. Here, we present gate-dependent multi-tip scanning tunneling microscopy transport measurements combined with photoemission experiments all performed in situ on pristine BiSbTe$_3$ thin films. To analyze the data, we develop a generic transport model including quantum capacitance effects. This approach allows us to quantify the gate-dependent conductivities, charge carrier concentrations, and mobilities for all relevant transport channels of three-dimensional topological insulator thin films (i.e., the two topological surface state channels, as well as the interior of the film). For the present sample, we find that the conductivity in the bottom surface state channel is minimized below a gate voltage of $V_{\text{gate}} = -34$ V and the top surface state channel dominates the transport through the film.

**RESULTS**

Angle-resolved photoemission spectroscopy

Figure 1a shows a schematic of the combined transport and photoemission measurement setup, which we use to analyze a $d = 10$ nm thin film of BiSbTe$_3$. ARPES results obtained at $h\nu = 8.4$ eV are shown in Fig. 1b, c. We find a linear dispersion of the TSS with

$\text{INTRODUCTION}$

Understanding the behavior of topological insulators under the influence of an electric field is of fundamental interest for the application of the unique electronic properties of their topological surface states (TSS) in future electronic devices. Among the most promising three-dimensional topological insulators (3D-TI) are the compounds Bi$_2$Se$_3$, Bi$_2$Te$_3$, and Sb$_2$Te$_3$ because of their pronounced band gap of up to 300 meV, which makes them applicable at room temperature. However, it has become clear that the aforementioned binary materials often suffer from unintentional doping by crystal lattice defects. This doping can shift the Fermi level $E_F$ into the bulk conduction/valence bands and may result in predominant bulk transport, bypassing the auspicious TSS. One way to reduce the bulk transport is to alloy different binary TIs into ternary or even quaternary alloys different binary TIs into ternary or even quaternary

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Received: 26 February 2018 Revised: 5 August 2018 Accepted: 7 August 2018

Published online: 17 September 2018
a filling level of the Dirac cone of $E_{\text{top}} = 250(20)$ meV, as determined from the intersection of the two dashed lines with respect to the Fermi energy. The cone slope corresponds to a Fermi velocity of $v_F = 5.6(2) \times 10^5$ m/s, in agreement with the literature. We further find that the position of the Dirac point coincides with the valence band edge (solid red line in Fig. 1c). A cut through the Dirac cone at $E_F$ (Fig. 1b) shows a circular Fermi surface with a Fermi radius of $k_F = 0.07(1)$ Å$^{-1}$. Spin-resolved energy distribution curves reveal a high spin polarization of the Dirac cone of up to 50%, consistent with the topological nature of the observed surface state (see Supplementary Note 1 and Supplementary Fig. 1). Using the Fermi wave vector $k_F$, we determine the carrier concentration in the TSS at the top surface of the sample, without a gate voltage applied, as $n_{\text{top}} = \frac{k_F^2}{2\pi} = 4(1) \times 10^{12}$ cm$^{-2}$ (ref. 9 and Supplementary Note 2). ARPES measurements were performed after the transport measurements to ensure the pristine sample quality throughout the measurements.

Transport measurements
Contacting the TI thin film with the tips of a four-tip STM results in a measurement geometry that resembles an in situ realization of a TI field-effect transistor. Four-probe $I/V$ measurements of the TI film at $V_{\text{gate}} = 0$ V, shown in Fig. 1d, reveal a linear current–voltage characteristic, corresponding to a sheet conductivity of $\sigma_{\text{total}} = 0.44(5)$ mS $\square^{-1}$. The error of $\sigma_{\text{total}}$ includes the positioning errors of the tips.

In the next step, we determine the TI film conductivity as a function of the gate voltage $V_{\text{gate}}$, resulting in the black squares in Fig. 2. In the data, we observe a transition between two distinct gating regimes: for gate voltages larger than $V_{\text{gate}} \approx -34$ V, the TI sheet conductivity shows a clear increase with $V_{\text{gate}}$, while for lower gate voltages, the conductivity saturates and forms a plateau at $\sigma_{\text{total}} = 0.34(5)$ mS $\square^{-1}$. The crucial advantage of the present study is the possibility to grow and characterize samples without breaking the vacuum. This is important because sample processing under ambient conditions, such as lithography, is reported to alter the electronic properties of TI samples and thus, the results of in situ photoemission measurements and ex situ transport measurements cannot be directly compared. In contrast, our seamless in situ approach allows a direct comparison of the respective results and enables the comprehensive analysis of our gate-dependent transport data.

Transport model
The conduction in a TI thin film can be described by three parallel conduction channels—the two TSS channels at the top and bottom surfaces of the film and one in the interior of the film, in the following referred to as “bulk channel”. Here, our gate-dependent transport measurements in combination with ARPES and a detailed transport model allow us to disentangle the contributions of the individual conduction channels. Specifically,
Where \( \sigma \) is the respective carrier mobilities, \( n \) is the carrier concentration without gating (i.e., \( V_{gate} = 0 \) V), and \( \mu \) is the corresponding gate-induced carrier concentrations, respectively. Note that there is a minimum carrier concentration in the TSS which is typically dominated by charge puddles.\(^{21}\) For the present sample, we assume a minimum charge carrier density of \( 0.5 \times 10^{12} \text{ cm}^{-2} \) based on the literature.\(^{12,21}\) We also show that varying this value does not have a large influence on the results obtained from applying the transport model to the present sample (see Supplementary Note 3, Supplementary Fig. 2 and Supplementary Table 1).

While \( n_{top}^0 \) is readily extracted from the ARPES measurements, \( n_{bot}^0 \) is difficult to access experimentally. For a freestanding topological insulator film and without gating, the position of the Fermi level relative to the Dirac point should be identical for the top and bottom TSS channels and correspondingly \( n_{bot}^0 = n_{top}^0 \). However, it has become clear that this is typically not the case in experiments.\(^{12,22}\) The different environments which the top and bottom TSS channels are exposed to result in different conductivities; e.g., at the bottom surface, the substrate may influence the carrier concentration in the TSS.\(^{16,18,19}\)

When gating the TI film, the carrier concentration in each of the three transport channels changes due to quantum capacitance effects, i.e., the gate-electric field penetrates through the TSS at the bottom surface of the film (see also Supplementary Note 4). These effects occur because, as charges are transferred to the TSS, higher energy levels must be populated due to the small density of states (DOS) in the TSS. Thus, part of the applied gate voltage is consumed by filling higher energy levels and, as a result, the gate effect is attenuated compared with a sample with a large DOS. In the corresponding equivalent circuit diagram, the quantum capacitance is described by an additional capacitance \( C_Q = e^2 n/E \) in series with the ordinary geometric capacitance, with \( n \) being the number of induced charge carriers on the capacitor plate and \( E \) the resulting shift of the filling level (see Supplementary Note 4 and Supplementary Fig. 3). For small changes in the carrier concentration, this expression can be approximated to \( C_Q \approx e^2 n_{bot}^0 / E \), where \( n \) is the DOS at the Fermi level.\(^{21,24}\) In the following, we lift this approximation for \( C_Q \) and implement an explicit treatment of the quantum capacitance in the form of the gate-dependent change of the TSS filling level. To include the quantum capacitance effect of the top and bottom TSS channels, we use a gating model given by the equivalent circuit diagram shown in Fig. 3a, after refs.\(^{15,21,24}\). In this description, we use capacitances per unit area and assume that the current injecting tips contact both TSS channels (close to ground potential) due to a finite conductance of the bulk channel.

**Gate-dependent carrier concentrations**

From the equivalent circuit model shown in Fig. 3a and charge conservation it holds\(^{15,21,24}\)

\[
\begin{align*}
n_{bot} &= -n_{gate} - n_{bot}^0, \quad \text{and} \quad n_{top} = n_{top}^0.
\end{align*}
\]

Using the definition of capacitance, the charge carrier densities on
the capacitors $C_{\text{gate}}$ and $C_{\text{Tl}}$ can be written as a function of the potential differences across them

$$n_{\text{gate}} = \frac{C_{\text{gate}}(E_{\text{gate}} - E_{\text{bot}})}{e^2}, \quad \text{and} \quad n_{\text{Tl}} = \frac{C_{\text{Tl}}(E_{\text{top}} - E_{\text{bot}})}{e^2}. \tag{3}$$

For the present sample, we determine $C_{\text{Tl}} = e_{\text{Tl}}/d = 8 \mu \text{cm}^{-2}$ from the TI film thickness $d$ and the dielectric constant $e_{\text{Tl}} = 100$. (Ref. 2) Note that the reported values for the dielectric constant of TIs fluctuate in the range $e_{\text{Tl}} \approx 50$–200 $e_{0}$. (Ref. 25) and that we have also investigated the dependence of our model on this parameter (see Supplementary Note 5 and Supplementary Fig. 4). Due to the linear dispersion of TSS, the filling level with respect to the Dirac point $E_{\text{top}}$ is furthermore related to the charge carrier density $n_{\text{top}}$ (for $V_{\text{gate}} = 0 \text{V}$) by

$$E_{\text{top}} = \sqrt{4\hbar^2 v_F^2 n_{\text{top}}^0} \equiv \sqrt{n_{\text{top}}} \cdot \tag{4}$$

and in the same way $E_{\text{bot}} = a\sqrt{n_{\text{bot}}}$ (see Supplementary Note 2 and refs. 9,12,26). A change in the carrier concentration $n_{\text{top}}^0 \rightarrow n_{\text{top}}^1 + n_{\text{top}}$ according to Eq. (4) therefore results in a change of the filling level $E_{\text{top}}$ given by $E_{\text{top}}^0 + E_{\text{top}} = a\sqrt{n_{\text{top}}^0 + n_{\text{top}}}.

From this, we obtain

$$E_{\text{top}} = a\left(\sqrt{n_{\text{top}}^0 + n_{\text{top}} - \sqrt{n_{\text{top}}^0}}\right) \tag{5}$$

and an equivalent expression for $E_{\text{bot}}$. Combining Eqs. (2), (3), and (5), we find the explicit equations

$$n_{\text{top}} \approx \left(\frac{C_{\text{top}}}{e^2}\right)^{-1} \left(-n_{\text{bot}} + \frac{C_{\text{Tl}}}{C_{\text{top}}} \left(\sqrt{n_{\text{top}}^0 + n_{\text{bot}} - \sqrt{n_{\text{top}}^0 + \sqrt{n_{\text{bot}}^0}}}\right)\right) \tag{6}$$

and

$$n_{\text{bot}} = \left(\frac{C_{\text{bot}}}{e^2}\right)^{-1} \left(-n_{\text{top}} + \frac{C_{\text{Tl}}}{C_{\text{bot}}} \left(\sqrt{n_{\text{bot}}^0 + n_{\text{bot}} - \sqrt{n_{\text{bot}}^0 + \sqrt{n_{\text{bot}}^0}}}\right)\right)^2 \tag{7}$$

(Supplementary Note 6). We evaluate Eqs. (6) and (7) numerically, which gives us $n_{\text{top}}$ and $n_{\text{bot}}$ as a function of $V_{\text{gate}}$ and the parameter $n_{\text{bot}}^0$, which is provided by ARCES. For a given value of $n_{\text{bot}}^0$, which we determine later by fitting the experimental data, we calculate the band bending in the TI film as a function of the applied gate voltage by modeling the TI film as a 10 nm thin small bandgap semiconductor ($E_{\text{gap}} = 260 \text{meV}$, according to our ARCES results) and solving Poisson’s equation (see Supplementary Note 7, Supplementary Fig. 5 and refs. 27–29). As a result, we obtain the gate-dependent total charge carrier density in the bulk channel $n_{\text{film}}^0 + n_{\text{film}}(V_{\text{gate}})$.

Before applying Eq. (1) to our data, note that Eqs. (6) and (7) are only valid if on each surface the Fermi level sits below the energy gap. The high DOS of the bulk bands corresponds to a large quantum capacitance of the respective surface and results in a complete screening of the gate electric field at this surface.24 For the present sample, this leads to a constant, gate-independent carrier concentration in the top TSS channel for gate voltages lower than $V_{\text{gate}} = -34 \text{V}$. Thus, further decreasing the gate voltage will lead to any further shift of the bands with respect to $E_F$. Combining the solution of Eqs. (6) and (7), when the Fermi level lies in the band gap, with the constant band positions once the bulk bands reach the Fermi level allows us to describe the entire range of experimental data, as shown in the following.

Disentanglement of the transport channels

We now turn to the quantitative analysis of the data in Fig. 2 using Eq. (1) in combination with $n_{\text{bot}}^0$, obtained from ARCES, the gate-dependent carrier concentrations $n_{\text{bot}}$, and $n_{\text{top}}^0$ determined by the band bending calculations. From a fit of Eq. (1) to the data, we determine the four remaining parameters of the transport model, i.e., $n_{\text{bot}}^0$, $\mu_{\text{bot}}$, $\mu_{\text{top}}$, and $\mu_{\text{film}}$. The results are $n_{\text{bot}}^0 = 1.7 \times 10^{12} \text{cm}^{-2}$, $\mu_{\text{bot}} = 184 \text{cm}^2\text{V}^{-1}\text{s}^{-1}$, $\mu_{\text{top}} = 578 \text{cm}^2\text{V}^{-1}\text{s}^{-1}$, and $\mu_{\text{film}} < 2 \text{cm}^2\text{V}^{-1}\text{s}^{-1}$. The resulting graph of Eq. (1) is shown as solid red line in Fig. 2. Additionally, we plot the individual contributions of the transport model $\sigma_{\text{bot}}$, $\sigma_{\text{top}}$, and $\sigma_{\text{film}}$. We find that the four parameters determined by the fit correspond to four specific signatures of the gate-dependent conductivity: (a) $n_{\text{bot}}^0$ is mainly determined by the gate voltage at which the kink in the conductivity occurs ($V_{\text{gate}} = -34 \text{V}$), (b) $\mu_{\text{bot}}$ is mainly determined by the conductivity value at the kink together with $n_{\text{bot}}^0 = 4 \times 10^{12} \text{cm}^{-2}$ obtained from ARCES, (c) $\mu_{\text{top}}$ is determined by the slope of the conductivity graph right of the kink, and (d) $\mu_{\text{film}}$ is determined by the slope of the graph left of the kink. At this point, we would like to stress that without the knowledge of $n_{\text{bot}}^0$, from the in situ ARCES measurements, the
fit of the transport model to the experimental data would be
underdetermined and would not result in a unique solution for
the fit parameters. Note also that the gating model reproduces
the experimentally observed negative curvature of the conductivity
graph above $V_{\text{gate}} = -34$ V, which is a direct result of the quantum
capacitance and in marked contrast to the expected linear graph if
only the bottom TSS channel would be affected by the gate.

Resulting from the fit of our transport model we find that for
gate voltages below $V_{\text{gate}} = -34$ V, the current through the sample
is almost exclusively transmitted by the top TSS channel. Beyond
this gate voltage the conductivity of the bottom TSS channel is
minimized because the Dirac point is located at the Fermi level, as
depicted in Fig. 3c. At this filling level and in the absence of
scattering, the conductance of the TSS is $2e^2/h$ (ref. 30). However, it
was recently shown that there is a significant amount of scattering
of the TSS at defects in the present samples. 31 In combination with
the large probe spacing in the present experiment (100 µm), this
results in a negligible amount of scattering due to the valence band edge at the bottom of the film
being located at the Fermi level. As we observe a constant conductivity
below $V_{\text{gate}} = -34$ V, we obtain a lower limit for the film mobility $\mu_{\text{film}} < 2$ cm$^2$ V$^{-1}$ s$^{-1}$. Our result for $\mu_{\text{film}}$ is two orders of magnitude smaller than the TSS mobility. In the literature, the bulk mobility is
reported to be typically one order of magnitude lower than the
conduction band mobility. 32 We explain our observed lower bulk mobility by the fact that our transport experiments were
performed at room temperature, resulting in an increased phonon scattering in the bulk compared with experiments performed at
low temperature. In contrast, the mobility in the TSS is reported to be
rather insensitive to phonon scattering. 33 The bulk charge
carrier densities obtained from our gate-dependent band bending
calculations agree with typical carrier densities reported in the
literature. 33 In combination with the estimated low mobility, this
results in a negligible film conductivity as evident in Fig. 2. We can exclude possible screening effects of the substrate and its interface
to the TI, as we have measured several samples of different
thicknesses and compositions on identically prepared substrates
which all show distinctly different gate-dependent conductivities
(see Supplementary Note 8, Supplementary Figs. 6–8 and
Supplementary Table 2).

In general, our transport model describes the experimental data
in Fig. 2 very well, with only small deviations between the
experimental and calculated graph when the filling in the bottom
TSS channel approaches the Dirac point. This observation can be
explained by a smooth transition between the gating regimes in the
experiments due to the room temperature conditions and the
resulting smeared-out Fermi edge. A width of the Fermi edge of
$\pm 2k_B T$, i.e., $E_{\text{F}} \pm 2k_B T$, translates to a broadening of the charge
carrier density $n_{\text{bot}}$ according to Eq. (4), and therefore after Eq. (2)
and (3), it leads to a broadening of $V_{\text{gate}}$ by $\pm 5$ V at room
temperature. This value fits the experimentally observed transition region in Fig. 2 reasonably well. Another effect which can
contribute to the smeared-out transition is local variations in the
band positions within the TI, e.g., due to charge puddles. 15

Figure 4a shows the disentangled gate-dependent valence and
conduction band positions which we calculate from the fitted
transport model after Eqs. (6) and (7). We find that while the
bottom TSS channel band positions $E_{\text{F}}^{\text{bottom}}$, $E_{\text{cb}}^{\text{bottom}}$ shift considerably as a function of gate voltage, the band positions in the top TSS channel changes only by an amount of $\Delta E < 50$ meV in the
experimentally applicable gate voltage range. This agrees with the
partial screening of the gate-induced electric field by the bottom
TSS channel (due to the quantum capacitance) and the larger
initial filling level of the top TSS channel, which results in a slower
change of the band positions as charges are induced, as a result of
the square-root dependence of the TSS filling level on the carrier
concentration (Eq. (4)). For $V_{\text{gate}} \leq -34$ V, the bulk valence band at the bottom of the film is located at the Fermi level and the band positions are constant as a result of the large DOS in this band.

While Fig. 3b, c showed only a qualitative sketch of the band
positions in the film, we can now calculate the exact band
positions from the fitted transport model. Figure 4b, c show the calculated depth-dependent profiles of the valence band edge $E_{\text{vb}}$
and conduction band edge $E_{\text{cb}}$ throughout the thin film at $V_{\text{gate}} = 0$ V and $V_{\text{gate}} = -34$ V, respectively. From these graphs, it is
evident that the Dirac point of the bottom TSS channel is shifted
to the Fermi level by gating, resulting in a minimal bottom TSS
channel conductivity at $V_{\text{gate}} = -34$ V, while at the top surface, $E_{\text{F}}$
lies within the bulk bandgap, with the largely filled TSS
dominating the conduction through the TI thin film. Furthermore,
itis evident that the bands show only a weak curvature, which is
the result of the long screening length compared with the film
thickness. 34
DISCUSSION
Our generic transport model shows that the influence of the gate voltage on the conductivity of each of the two TSS channels in a TI thin film can vary from strong to negligible, depending on the alignment of the bulk bands with respect to $E_F$ at each surface. In detail, we find three possible cases: (1) If the Dirac points of both TSS channels are located well within the bandgap, the influence of the gate voltage is strong and controls the charge transport through both TSS channels.\textsuperscript{15,24} (2) If the gate shifts the bands such that the Fermi level enters at one or both surfaces into the bulk bands (as it was observed in refs. \textsuperscript{12,13}), both TSS channel fillings become independent of the gate voltage because the high DOS of the bulk bands at this surface screens the gate electric field completely. This effect explains also the often-reported negligible gate-dependence in first-generation TI samples, where strong doping resulted in $E_F$ being located well within the valence/conduction bands in the range of applicable gate voltages.\textsuperscript{1,29} Our transport model can capture all these cases of band alignments quantitatively. The comparison between the model and the experiment would greatly benefit from facilitating gate-dependent ARPS measurements, which would allow to determine $n_{opt}(V_{gate})$ directly. However, such experiments were not possible in the present study because the gate electric fields need to be well screened to not influence the ARPS measurements, and this would have required elaborate changes to the experimental setup.

In conclusion, by combining in situ gate-dependent transport measurements with photoemission spectroscopy, we are able to disentangle the transport through all of the conduction channels of a Bi$_2$SbTe$_3$ thin film, namely the top and the bottom TSS channels, as well as the interior of the film. The use of a generic transport model, which includes gate-dependent quantum capacitances, gives access to the carrier concentrations and respective mobilities in each of the three channels. Application of the model to the experimental data shows that for the present sample the conductivity in the bottom TSS channel is minimized below a gate voltage of $V_{gate} = -34\, V$, resulting in the top TSS channel dominating the transport through the film. The reason for this behavior is a high DOS in the bulk bands of the TI, which prevents a gate-induced shift of the bulk band edges past the Fermi energy, in combination with the Dirac point being located at the bulk valence band edge in the present sample. The demonstrated experimental techniques and data analysis procedures are generic and allow to determine the detailed transport properties of pristine samples with short turnaround times.

METHODS
Sample preparation
We prepared a $d = 10\, \text{nm}$ thin film of (Bi$_{0.53}$Sb$_{0.47}$)$_2$Te$_3$ on a silicon-on-insulator (SOI) wafer by molecular beam epitaxy. The SiO$_2$ thickness is $300\, \text{nm}$, while the template layer on which the TI film is grown consists of undoped Si(111) with $70\, \text{nm}$ thickness. For further details see Supplementary Note 9.

Angle-resolved photoemission spectroscopy
ARPES measurements were performed at 30 K with a MBS A-1 hemispherical energy analyzer that was set to 40 meV energy resolution. Monochromatic VUV light with $h\nu = 8.4\, \text{eV}$ came from a microwave-driven Xe discharge lamp. Fermi surface maps were obtained by rotating the sample around the in-plane sample axis that is parallel to the analyzers entrance slit.

Four-probe measurement
For transport measurements, we use an mProbes room-temperature four-tip STM.\textsuperscript{30} This instrument allows individual positioning of the tips on the sample surface under scanning electron microscope monitoring. To measure the sample conductivity, the tips are lowered out of tunneling contact until a soft but stable contact to the sample surface is formed. Details on the measurement technique can be found, e.g., in ref. \textsuperscript{17}. For all STM experiments, we used electrochemically etched tungsten tips.

DATA AVAILABILITY
Data within the manuscript, and its Supplementary Information is available from the corresponding author upon reasonable request.

ACKNOWLEDGEMENTS
We gratefully acknowledge Helmut Stollwerk and Franz-Peter Coenen for technical assistance.

AUTHOR CONTRIBUTIONS
F.L., S.J., M.E., T.H., E.M., M.L., and N.v.d.D. performed the experiments and evaluated the data. F.L., V.C., and B.V. designed the transport experiment. M.E., T.H., E.M., L.P., and C.M.S. designed the photoemission experiment. M.L., P.S., D.R., N.v.d.D., G.M., and D.G. developed and fabricated the samples. The manuscript was written by F.L., S.J., M.E., T.H., M.L., F.S.T., and B.V. All authors discussed and commented on the manuscript.

ADDITIONAL INFORMATION
Supplementary information accompanies the paper on the npj Quantum Materials website (https://doi.org/10.1038/s41535-018-0116-1).

Competing interests: The authors declare no competing interests.

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

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