Mapping the chemical potential dependence of current-induced spin polarization in a topological insulator

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We report electrical measurements of the current-induced spin polarization of the surface current in topological insulator devices where contributions from bulk and surface conduction can be disentangled by electrical gating. The devices use a ferromagnetic tunnel junction (permalloy/Al₂O₃) as a spin detector on a back-gated (Bi,Sb)₂Te₃ channel. We observe hysteretic voltage signals as the magnetization of the detector ferromagnet is switched parallel or antiparallel to the spin polarization of the surface current. The amplitude of the detected voltage change is linearly proportional to the applied dc bias current in the (Bi,Sb)₂Te₃ channel. As the chemical potential is tuned from the bulk bands into the surface state band, we observe an enhancement of the spin-dependent voltages up to 300% within the range of the electrostatic gating. Using a simple model, we extract the spin polarization near charge neutrality (i.e., the Dirac point).

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I. INTRODUCTION

The combination of strong spin-orbit coupling, time-reversal symmetry, and inversion symmetry is known to create gapless helical Dirac surface states that lie within the bulk band gap of narrow band gap semiconductors such as the Bi-chalcogenides. These three-dimensional (3D) topological insulators [1–3] have begun to attract significant contemporary interest for spintronics since the spin-momentum locking in the surface state endows an inherent spin polarization to surface-state charge currents [4,5]. This has motivated several recent experiments on topological spintronics. Spin-transfer torque has been demonstrated in bilayers of ferromagnets and topological insulators [6–8], with record values of the spin torque ratio at room temperature in Bi₂Se₃ [6] and at low temperature in (Bi,Sb)₂Te₃ [7]. Spin Hall angles have also been measured through the inverse spin Hall effect in topological insulators, created by the injection of a spin current from a metallic ferromagnet through spin pumping [9–11] and through spin-polarized tunneling [12].

The spin polarization of the surface states of topological insulators has been measured in extensive spin- and angle-resolved photoemission spectroscopy studies [13–16]. However, direct electrical measurement, relevant for developing spintronic devices, are just beginning to emerge and derive from a well-established methodology in spintronics: the essential idea is to use a ferromagnetic contact as a voltage probe of the spin-dependent electrochemical potential. As the relative orientation of the current spin polarization and ferromagnet magnetization changes, so does the measured voltage [17]. This scheme was first used to clearly demonstrate spin-momentum locking using ferromagnetic tunnel contacts on Bi₂Se₃ transport channels, albeit with the chemical potential of Bi₂Se₃ in the bulk conduction band and at temperatures below 150 K [18]. More recent experiments have extended such measurements to room temperature in Bi₂Se₃ [19] and to topological insulators with reduced bulk conduction [20–22]. Further, a recent study used spin-polarized tunneling in ferromagnet/insulator/topological insulator junctions to map out the energy-dependence of the effective spin polarization in Bi₂Se₃ [12]. Here, we demonstrate an experiment that is complementary to these prior studies. We show that the standard three-probe spin potentiometry scheme using a magnetic tunnel junction (MTJ) [17,18] can be effectively applied to electrically gated topological insulator devices, thus yielding insights into the chemical potential dependence of the spin polarization of the surface state current. In our measurements, we map out the spin-dependent voltage as the chemical potential is tuned from the bulk conduction band through the Dirac point and into the valence bulk band. We find that the spin-dependent voltages are enhanced as the chemical potential is moved away from the bulk conduction/valence bands into the surface state band. We also show that we do not observe possible contributions of the opposite spin polarization from Rashba spin-split bands within the range of the electrostatic gating.

II. SAMPLE SYNTHESIS AND DEVICE FABRICATION

(Bi,Sb)₂Te₃ thin films were grown on SrTiO₃ substrates by molecular beam epitaxy (MBE) [23]. We used a Bi : Sb ratio of ∼1 : 1 to place the chemical potential into the bulk band gap. The samples had film thicknesses of 7 nm (S1), 8 nm (S2), and 6 nm (S3); this minimizes contributions from unpolarized bulk carrier conduction and allows the tuning of the electron chemical potential of both the bottom and top surfaces by back gating. We note that the film thicknesses are above the limit where Dirac point is disrupted by hybridization of the top and bottom surface states [16,24,25].

For clean measurements of the surface state spin polarization, we developed a device geometry that restricts all measurements to the top surface [Fig. 1(a)]. We first define a cross-shaped (Bi,Sb)₂Te₃ channel by standard photolithography followed by Ar plasma etching. A bilayer photoresist window of circular or rectangular shape is then patterned at the center of the cross-shaped channel. The bare surface of the...
topological insulator is cleaned using a low-power Ar plasma etch (15 W for 20 s) that removes possible native oxides and photoresist residues along with ~1 nm (Bi,Sb)2Te3. Following the sputter deposition of an Al2O3 seed layer (~0.3 nm), we use atomic layer deposition (ALD) for conformal deposition of an Al2O3 layer (2.1 nm for devices on S1 and 1.8 nm for devices on S2) over the topological insulator surface. A Py layer (20 nm) and a thin Au capping layer (5 nm) are then deposited by e-beam evaporation, followed by a lift-off process. Finally, the top of the MTJ is extended to two Au contact pads over a 60-nm-thick Si3N4 layer to isolate the MTJ from the topological insulator channel as shown in Figs. 1(b) and 1(e). Each sample has multiple devices with various MTJ areas. The five devices studied in this paper are listed in Table I.

For microstructural analysis, a device was cross sectioned using a focused ion beam (FIB, FEI Quanta 200 3D) and then imaged using high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) [Figs. 1(d) and 1(e)]. For STEM imaging, an aberration-corrected (CEOS DCOR probe corrector) FEI Titan G2 60-300 S/TEM equipped with a Schottky X-FEG gun was operated at 200 kV with a probe convergence angle of 16 mrad. Additionally, the Al2O3 layer is identified as being amorphous by conventional TEM imaging (not shown), using an FEI Tecnai G2 F30 at 300 kV.

III. RESULTS AND DISCUSSIONS

A. Ambipolar transport of (Bi,Sb)2Te3

We first discuss the back-gate-voltage (VG) dependence of the channel resistance and the Hall resistance. The former is measured through the two-terminal channel voltage between leads 2 and 4 and the latter between 3 and 5 is measured while a dc current of 1 μA flows between leads 2 and 4 [see Fig. 1(a) for lead configuration]. The channel between two Ti/Au leads (between 2 and 4 as well as between 3 and 5) is 300 μm long by 200 μm wide, and the contact resistance of each lead is ~100 Ω. The Hall voltage was measured as a function of applied perpendicular-to-plane magnetic field of 10 kOe. The results show the typical signatures of ambipolar transport in a topological insulator: the change of carrier type from electrons to holes as well as a peak in channel resistance near the charge neutrality point (at VG = 13 V for S1 and at VG = 10 V for S2) [Figs. 2(a) and 2(b)]. Carrier densities at each end of the applied gate-voltage range were obtained to be $n_e = 3.53 \times 10^{13} (2.44 \times 10^{13})$ cm$^{-2}$ at VG = 140 V and $n_p = 1.91 \times 10^{14} (1.69 \times 10^{14})$ cm$^{-2}$ at VG = −140 V for S1 (S2), respectively [Fig. 2(c)]. These values give an idea of the range of the chemical potential tuning. We believe that we were able to change the chemical potential across the bulk band gap from a position near the bottom of the conduction band (at 140 V) to a position below the top of the valence band (at −140 V).

B. Weak antilocalization of (Bi,Sb)2Te3

To further understand the surface and bulk conduction in the (Bi,Sb)2Te3 films with respect to the gate voltage, we carried out magnetotransport measurements using the same wiring configuration. Near 2 K, the magnetoresistance of a topological insulator channel shows a sharp cusp near zero magnetic field as shown in the inset to the Fig. 2(d). This suppression of resistance near zero magnetic field can be explained by weak antilocalization, typically seen in conventional 2D metals with strong spin-orbit coupling [26] as well as in 3D topological

TABLE I. List of devices with different MTJ areas, MTJ shapes, (Bi,Sb)2Te3 samples, thicknesses of (Bi,Sb)2Te3 film, and thicknesses of Al2O3 tunnel barrier.

<table>
<thead>
<tr>
<th>Device</th>
<th>MTJ Area (μm²)</th>
<th>MTJ Shape</th>
<th>Sample (Bi,Sb)2Te3 t (nm)</th>
<th>Al2O3 t (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dev1</td>
<td>502π</td>
<td>circle</td>
<td>S1</td>
<td>7</td>
</tr>
<tr>
<td>Dev2</td>
<td>1002π</td>
<td>circle</td>
<td>S1</td>
<td>7</td>
</tr>
<tr>
<td>Dev3</td>
<td>50 × 50</td>
<td>square</td>
<td>S1</td>
<td>7</td>
</tr>
<tr>
<td>Dev4</td>
<td>10 × 80</td>
<td>rectangle</td>
<td>S2</td>
<td>8</td>
</tr>
<tr>
<td>Dev5</td>
<td>40 × 80</td>
<td>rectangle</td>
<td>S3</td>
<td>6</td>
</tr>
</tbody>
</table>
Insulators that have a nontrivial \( \pi \) Berry’s phase [13]. The quantum corrections to the diffusive magnetoconductance are given by Ref. [27]

\[
\sigma(H) - \sigma(0) = \alpha e^2 / 2\pi^2 \hbar \left[ \psi \left( \frac{1}{2} + \frac{\hbar c}{4e^2 \sigma H} \right) - \ln \left( \frac{\hbar c}{4e^2 \sigma H} \right) \right].
\]  

(1)

Here, \( \sigma \) is the 2D channel conductivity and can be determined from the 2D channel resistivity \( \rho_{xx} \) and 2D Hall resistivity \( \rho_{xy} \) via \( \sigma = \rho_{xx}/(\rho_{xx}^2 + \rho_{xy}^2) \). Also, \( e, \hbar, \psi(x), \alpha, \) and \( \sigma \) are the electronic charge, the Planck’s constant, the digamma function, the prefactor, and the coherence length, respectively.

C. Tunnel junction properties

The RA product of the tunnel junctions is in the range \( 10^8 - 10^9 \) MΩ μm². We observe qualitatively similar behavior of the gate-voltage dependence, the tunnel junction characteristics, and the spin-dependent voltages from all the devices. Typically, the temperature dependence of the zero-bias junction resistance shows an insulating behavior [Fig. 3(a)], while the channel resistance of the topological insulator shows metallic behavior below 100 K [Fig. 3(a), inset]. The junction resistance as a function of measured dc voltage (\( V_{dc} \)) across an MTJ shows an anomaly at zero-bias [Fig. 3(b)], commonly seen in MTJs [31]. We also observed an asymmetry in the junction resistance between positive and negative voltages [Fig. 3(b)].

Interestingly, the asymmetric behavior qualitatively follows the gate-voltage dependence of the channel resistance [Fig. 2(a)]. The asymmetry of the junction resistance as a function of bias voltage has been reported in previous measurements of magnetic tunnel junctions and attributed to effects such as interfacial roughness and impurities in the tunnel barrier [32]. The systematic changes of the asymmetric behavior that we observe as a function of gate voltage suggest the asymmetry could have a different origin: we speculate that
it arises from probing of the density of states of the topological insulator top surface via tunneling between the $V_{dc}$-tuned metal and the $V_{G}$-tuned topological insulator. This interpretation is consistent with recent tunneling spectroscopy measurements of Bi$_2$Se$_3$/MgO/CoFeB devices [12].

### D. Spin-dependent voltages from topological insulator spin polarization

Our three-terminal potentiometric measurement scheme follows the proposal by Hong et al. [17]. We use an Al$_2$O$_3$/Py junction for the ferromagnetic electrode while a dc current flows through a topological insulator channel with an in-plane magnetic field perpendicular to the current direction. When the direction of the Py magnetization switches from parallel to antiparallel to the current-induced spin polarization of the topological insulator surface state, we observe steplike changes in the detected voltage, resulting in hysteretic spin-dependent voltages associated with the coercive field (~100 Oe) of the Py layer. For positive currents as shown in Figs. 4(a) and 4(b), the relative direction between the Py magnetization and the topological insulator spin polarization determines the positive change in the detected voltage $\Delta V = [V(M) - V(-M)]$. For reversed currents [Figs. 4(c) and 4(d)], the direction of topological insulator spin polarization is opposite to the case of positive currents, so that the change in the detected voltage $\Delta V$ becomes negative. Figures 4(e) and 4(f) show that the measured voltage change is linearly proportional to the bias current. This linear relation can be expressed by

$$\Delta V = I R_B (p - m),$$  \hspace{1cm} (2)

where $R_B$, $p$, and $m$ represent ballistic resistance, degree of the spin polarization per unit current in the topological insulator channel, and effective magnetic polarization of ferromagnet, respectively [17]. Depending on the position of the chemical potential tuned by the electrical gating, the measured spin-dependent voltages may originate not only in the topological insulator surface states but also in the Rashba spin-split bands. Equation (2) is applicable to the spin-dependent voltages from combined conduction channels including the surface states, the Rashba spin-split bands, and the unpolarized bulk bands in both ballistic and diffusive limits.

Most of our measurements of gate-voltage dependence of spin-dependent voltages described below were performed at 1.8 K. We also studied the temperature dependence of the spin-dependent voltages at a fixed voltage ($V_{dc} = 0$ V) and found that the amplitude of the hysteretic spin-dependent voltages decreases as the temperature is increased, vanishing at about 20 K. This is similar to previous observations [22]. We note however that the reason for this decrease is likely not intrinsic but probably stems from an imperfect tunnel barrier. For instance, in other work [12,18], the use of a higher-quality tunnel barriers allows measurement of spin-dependent signals up to significantly higher temperatures.

### E. Gate-voltage dependence of spin-dependent voltages

We now measure the hysteretic spin-dependent voltages while changing the chemical potential across the bulk band gap. When the chemical potential is placed in the bulk band gap, the observed spin-dependent voltages are two to three times larger. Using two devices (Dev3 from S1 and Dev4 from S2), multiple measurements of magnetic field sweeps, repeated at least eight times, were carried out every 10 V from 140 V to $-140$ V. Figures 5(a) and 5(b) show the hysteretic spin-dependent voltages at some of the $V_G$’s among the multiple measurements. As shown in Figs. 5(c) and 5(d), the magnitude of the resulting voltage change $\Delta V$ is maximal near the charge neutrality point ($V_G = 13$ V for S1 and $V_G = 10$ V for S2).

We do not observe any sign change in $\Delta V$ throughout the $V_G$ range, suggesting that with a fixed current bias, the carrier type change from $n$ to $p$ does not flip the direction of the measured spin polarization. This observation is fully consistent with dominant contributions to the spin polarization originating from the helical Dirac surface states since we use a dc current for the measurement. For a given crystal momentum, the spin polarization of holes is opposite to that of electrons. However, with a fixed dc current, the propagation...
FIG. 4. (Color online) Detected spin-dependent voltages with positive dc bias current (a) from Dev4 [3 \(\mu\)V (red) and 1 \(\mu\)V (green)] and (b) from Dev2 [30 \(\mu\)V (red), 10 \(\mu\)V (gray), and 3 \(\mu\)V (green)]. Reversed spin-dependent voltages with negative bias currents (c) from Dev4 [−4 \(\mu\)V (blue) and −1 \(\mu\)V (cyan)] and (d) from Dev2 [−30 \(\mu\)V (blue)]. The schematics depict spin polarization (red arrow) induced by electron current (black arrow) on a surface of a topological insulator channel and the magnetization of the ferromagnetic layer (white arrow). The voltage change \(\Delta V\) as a function of bias current (e) from Dev4 and (f) from Dev2 shows a linear relationship. The black line is a linear fit to data shown as red circles with error bars that represent the standard deviations over multiple measurements. Inset in (e) illustrates the measurement setup. All data were taken at 1.8 K with \(V_G = 0\) V.

Direction of holes (below the Dirac point) is the reverse of the propagation of electrons (above the Dirac point). Therefore, the opposite spin polarization and the opposite propagation direction between electrons and holes make the sign of the detected spin-dependent voltages remain the same above and below the Dirac point [12]. We also note that our observations do not reveal any significant contributions from coexisting Rashba states, which would have resulted in the opposite behavior due to the spin polarization being opposite to that of topological surface states [17].

For 3D bulk conduction, \(k_F = \sqrt{3\pi^2 n}\) where \(n\) is the 3D carrier density from all the bulk modes. However, with the chemical potential in the bulk bands, surface conduction may also coexist along with the bulk conduction, with an associated \(k_F = \sqrt{4\pi n_s}\) where \(n_s\) is the 2D carrier density of a surface conduction channel [33]. Thus, a proper accounting of the distribution of current between bulk and surface conduction channels is actually needed to interpret experimental data when the chemical potential lies in the bulk bands.

Our studies of ambipolar transport and weak antilocalization at different gate voltages suggest that, near the charge neutrality point, the bulk is depleted and a current flows only through the surface states. In this case, we can calculate \(k_F\) using the lowest carrier density near the charge neutrality point (7.72 \(\times\) 10\(^{12}\) cm\(^{-2}\) for Dev3 and 4.13 \(\times\) 10\(^{12}\) cm\(^{-2}\) for Dev4); this corresponds to the residual carrier density from the puddles of electrons and holes created by disorder [33,34]. For the surface state, we determine \(k_F = \sqrt{4\pi n_s}\) = 0.070 Å\(^{-1}\) for Dev3 and 0.051 Å\(^{-1}\) for Dev4. Using \(P_{FM} \approx 0.51–0.63\) for a 20 nm thick Py layer [35,36] and the maximal \(\Delta V\) near the charge neutrality point, we estimate the spin polarization of the topological insulator surface state \(P_{TI} = 0.42 \pm 0.15\) for Dev3 and 0.78 \pm 0.26 for Dev4. These values are consistent within the error bars and comparable to theoretically calculated values.
FIG. 5. (Color online) Detected hysteretic spin-dependent voltages with up sweep (red) and down sweep (black) of in-plane magnetic field with different gate voltages (a) from Dev3 and (b) from Dev4. Observed $\Delta V$ with respect to $V_G$ is plotted in (c) for Dev3 and in (d) for Dev4. The error bars represent the standard deviations over multiple measurements. All data were taken with fixed currents of 30 $\mu$A for Dev3 and 2 $\mu$A for Dev4 at 1.8 K.

of 0.5 [37] or $2/\pi$ [17]. We note that the estimated numbers are lower bounds since the effective magnetic polarization $P_{FM}$ could have lower values for nonideal Al$_2$O$_3$/Py junctions.

IV. CONCLUSIONS

In conclusion, we have carried out electrical measurements of the spin polarization of the surface currents in electrically gated (Bi$_x$Sb)$_2$Te$_3$ devices as a function of the chemical potential. We estimate the spin polarization of the topological surface state to be 0.42 ± 0.15 and 0.78 ± 0.26 from two devices. Larger spin-dependent voltages were detected as the chemical potential approaches the Dirac point. Our results suggest that to achieve the maximum spin-dependent voltages from 3D topological insulators it is important to avoid the effects from the bulk bands, such as the parallel conduction of the unpolarized bulk carriers or the opposite spin polarization by Rashba spin-orbit interaction. Also, the results point toward a new strategy for electrical control of the magnitude of spin-dependent voltages from the current-induced spin polarization for potential topological spintronics, using gate-tunable topological insulator films.

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