Study of Surface Spin-Polarized Electron Accumulation in Topological Insulators Using Scanning Tunneling Microscopy

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Received 6 Dec 2019, revised 22 Jan 2020, accepted 23 Jan 2020, published 28 Jan 2020, current version 2 Mar 2020.

Abstract—We report the results of scanning tunneling microscopy experiments using iron-coated tungsten tips and current-carrying bismuth selenide (Bi\textsubscript{2}Se\textsubscript{3}) samples. Asymmetry in tunneling currents with respect to the change in the direction of bias currents through Bi\textsubscript{2}Se\textsubscript{3} samples has been measured. It is argued that this asymmetry is the manifestation of surface spin-polarized electron accumulation caused by the 90° spin–momentum locking in the topologically protected surface current mode. It is demonstrated that the manifestation of surface spin-polarized electron accumulation is enhanced by tin doping of Bi\textsubscript{2}Se\textsubscript{3} samples. Furthermore, we noted the appearance of spin-dependent density of states in current-carrying Bi\textsubscript{2}Se\textsubscript{3} samples.

Index Terms—Spin electronics, topological insulators, scanning tunneling microscopy, potentiometry.

I. INTRODUCTION

Topological insulators are currently a very active area of research in physics in general, and spintronics, in particular, due to their unique physical properties [Hasan 2010; Moore 2010; Qi 2011; Ando 2013] and promising engineering applications [Mellnik 2014; Mahendra 2018; Claro 2019]. Topological insulators have a bulk bandgap like an ordinary insulator (or semiconductor) and conducting surface states topologically protected by time-reversal symmetry. These surface-conducting states exhibit 90° locking [Hasan 2010] between the electron spin and its momentum caused by very strong spin-orbit interaction in these materials. This 90° locking results in surface accumulation of spin-polarized electrons when a bias current flows through the topological insulator.

In this letter, we report the experimental study of these surface spin-polarized electron accumulations by using scanning tunneling microscopy (STM) with iron-coated tungsten tips which have been previously used in the STM study of different problems [Kleiber 2002; Bode 2005; Hanke 2005; Tamada 2005; Clavero 2010]. In our experiments, by using iron-coated tips, it was found that there is a change (i.e., asymmetry) in the tunneling current with respect to the change in the direction of the bias current through the topological insulator. It can be reasoned that this asymmetry is caused by the following two factors. The first is the change in the spin orientation of surface electrons caused by the change in the bias current direction. The second is the spin-dependent density of states of the iron-coated tungsten tip. Thus, it can be concluded that the above-mentioned asymmetry reveals local surface accumulation of spin-polarized electrons caused by the 90° spin–momentum locking.

Our experiments were performed using molecular-beam epitaxy (MBE) grown Bi\textsubscript{2}Se\textsubscript{3} samples. These are binary compounds that represent the second-generation of topological insulator materials [Hasan 2010] with a relatively large bulk bandgap of around 0.3 eV and the simplest (almost ideal) surface band structure with a single Dirac cone for the (topologically protected) conducting surface mode. In Bi\textsubscript{2}Se\textsubscript{3} samples, the unintentional bulk conductivity results in bulk currents, which obscure the surface conducting states [Hsieh 2009; Zhang 2012]. It has been shown that chemical doping of intrinsic topological insulators with such elements as calcium [Hor 2009; Hsieh 2009] or tin [Jaworski 2009; Zhang 2012] moves the bulk Fermi level into the bulk bandgap. This results in reduced conduction due to bulk states. In our experiments, we used Bi\textsubscript{2}Se\textsubscript{3} samples with different levels of tin (Sn) doping to reduce bulk conductivity. It was noted that the increase in tin doping levels results in the increase in tunneling current asymmetry with respect to the direction change of the bias current through the samples. The latter indicates the enhanced manifestation of the topological surface mode. It was also found that the current flow through the Bi\textsubscript{2}Se\textsubscript{3} samples results in the appearance of spin-dependent density of states in the samples. This was revealed by spin-polarized electron tunneling from the iron-coated tungsten tips to the samples. The appearance of this spin-dependent density of states in samples may be the result of inclined (i.e., not horizontal) crossing of the Dirac cone by the surface Fermi level in the presence of the bias current through Bi\textsubscript{2}Se\textsubscript{3} samples.

The STM technique has been extensively used for the study of physical properties of topological insulators [Cheng 2010; Romanowich...
2013; Zhang 2013]. This is because it is very local (nanoscale) in nature, which is its clear advantage in comparison with other electrical and optical measurements [Hasan 2010; Moore 2010]. The contributions of this letter are the local STM measurements of surface spin-polarized electron accumulations in Bi$_2$Se$_3$ samples with various tin doping levels by using iron-coated tips.

II. TECHNICAL DISCUSSION

Our STM studies were performed in a two-chamber Omicron ultrahigh vacuum (UHV) STM system at room temperature, using the experimental setup schematically shown in Fig. 1. The current source shown in this figure was used to provide the desired bias current $I_b$ through the Bi$_2$Se$_3$ sample, whereas the voltage source $V_s$ was used to apply the desired tunneling voltage between the STM tip and the sample. The bias current results in a voltage drop $V(x, y)$ (see Fig. 1) along the sample, and this complicates the application of the proper tunneling voltage ($V_{gap}$) between the tip and the sample. This problem was solved by developing the special potentiometry technique [Xie 2017], and thereafter, it was used in the STM study of the spin Hall effect [Xie 2018a, 2018b, 2018c]. Potentiometry adjusts the voltage $V_{in}$ appropriately to compensate for the voltage drop $V(x, y)$ and hence ensures that $V_{gap} = V_s$ even in the presence of bias currents through the sample.

The iron-coated tungsten tips were fabricated in UHV by using e-beam deposition of iron on clean tungsten tips. Scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDS) of the tip used for our experiments confirmed the presence of iron at the front end, as shown in Fig. 2.

The Bi$_2$Se$_3$ thin films with the thickness of about 400 nm were grown by MBE on (001) GaAs substrates. The growth proceeded by using a substrate temperature of 260 °C and an appropriate relative flux ratio of selenium to bismuth. Along with intrinsic Bi$_2$Se$_3$ samples, three tin-doped samples were grown epitaxially with different levels of tin doping. Sn doping was accomplished using a molecular beam of elemental tin whose flux was calibrated using a Bayard–Alpert ionization gauge. These three Sn-doped samples have their stoichiometry of the form (Bi$_{1-x}$Sn$_x$)$_2$Se$_3$ with $x$ being equal to 0.0073, 0.0216, and 0.049. Below, these samples shall be referred to as samples A, B, and C. The stoichiometry was determined by calibration of the tin flux measurements ($\Phi_{Sn}$) relative to those of pure bismuth ($\Phi_{Bi}$) prior to growth. The relative tin concentration was then obtained in the standard manner as $\Phi_{Sn}/(\Phi_{Sn} + \Phi_{Bi})$. These films were cut to 0.5 cm in width and 1 cm in length. To achieve clean surfaces of these Bi$_2$Se$_3$ samples for scanning, they were cleaved in vacuum and then placed in the UHV STM chamber.

Before conducting the experiments with current-carrying samples, STM images of the atomic structure of the Bi$_2$Se$_3$ sample surfaces were obtained along with spectroscopy measurements of the Dirac cone of the topologically protected surface conducting mode. Typical surface morphology for intrinsic (undoped) Bi$_2$Se$_3$ samples is shown in Fig. 3(a). Typical spectroscopy curves for Bi$_2$Se$_3$ samples are shown in Fig. 3(b), and are consistent with measurements previously reported [Romanowich 2013]. Note that the Dirac point corresponds to the minimum of the $dI/dV$ curve. Measurements shown in Fig. 3(b) reveal the shifting of the Dirac point toward zero tunneling voltage with the increase in the doping levels.

Next, the STM experiments with current-carrying Bi$_2$Se$_3$ samples were performed. As the initial step, a dc bias current through the sample in one direction was introduced, and the constant tunneling current mode of the STM tip was used to determine the achievement of thermal equilibrium. Then, the bias voltage ($V_{bias}$) and tunneling current ($I_{t}^{-}$) were adjusted to desired values. Subsequently, the constant tunneling current feedback for the STM tip was turned off, the direction of the bias current was abruptly changed to the opposite direction, and the tunneling current ($I_{t}^{-}$) was measured as a function of time. The superscript arrows in the above-mentioned notations refer to the direction of the bias current through the samples.
samples. The appearance of the tunneling currents measured for the samples (i.e., \( I_t \approx 1 \)). This implies that in the case of tungsten tips, tunneling currents are even-symmetric functions of bias currents.

Then, the STM experiments were conducted by using iron-coated tungsten tips. They were performed for a bias current of 20 mA through the Bi\(_2\)Se\(_3\) samples and tunneling currents (\( I_t \)) of about 100 pA. It was found that for iron-coated tips, the tunneling current was not an even-symmetric function of the bias current. The asymmetry was characterized by the ratio \( I_t^-/I_t^+ \) of the tunneling currents measured for the opposite direction of the bias current. The experimentally measured asymmetry of the tunneling currents for different values of tunneling voltages is presented in Fig. 4(a) for the intrinsic Bi\(_2\)Se\(_3\) sample and in Fig. 4(b)–(d) for tin-doped Bi\(_2\)Se\(_3\) samples A, B, and C, respectively.

First, the above-mentioned STM experiments were performed with tungsten (not iron-coated) tips with a bias current of 20 mA, and no significant changes in tunneling currents were detected upon changing the direction of bias currents through the Bi\(_2\)Se\(_3\) samples (i.e., \( I_t^-/I_t^+ \approx 1 \)). This implies that in the case of tungsten tips, tunneling currents are even-symmetric functions of bias currents.

It is important to note that, since no asymmetry \( I_t^-/I_t^+ \) in tunneling currents was detected in the case of tungsten tips, the reported asymmetry in Fig. 4 can be attributed to the spin-dependent tunneling of electrons between the Fe-coated tip and current-carrying Bi\(_2\)Se\(_3\) sample. This clearly reveals the spin sensitivity of Fe-coated tips.

In Fig. 4(a)–(d), the bottom curves (with \( I_t^-/I_t^+ < 1 \)) correspond to the polarity of tunneling voltages for which electrons tunnel from the current-carrying Bi\(_2\)Se\(_3\) samples to the iron-coated tip. On the other hand, the top curves (with \( I_t^-/I_t^+ > 1 \)) correspond to the opposite polarity of tunneling voltages for which spin-polarized electrons tunnel from the iron-coated tip to the current-carrying sample. The bottom curves reveal the surface spin-polarized electron accumulations caused by the 90° locking between momentum and spin of electrons of the topologically protected surface conducting mode in Bi\(_2\)Se\(_3\) samples. Indeed, this locking results in the reversal of electron spin orientation upon reversal of the bias current direction. This spin reversal leads to the change in tunneling currents due to the spin-dependent density of states of the iron-coated tip.

The top curves in Fig. 4(a)–(d), corresponding to the spin-polarized electrons tunneling from the iron-coated tip, reveal the spin-dependent density of states in current-carrying Bi\(_2\)Se\(_3\) samples. The appearance of this spin-dependent density of states may be the result of inclined (not horizontal) crossings of the Dirac cone by surface Fermi levels [Hus 2017; Siu 2018]. These inclined crossings are caused by bias surface currents in Bi\(_2\)Se\(_3\) samples. These bias surface currents are increased with the increase in tin doping levels of the sample. The latter one results in larger slope angles of surface Fermi levels, which leads to the increase in the spin-dependent density of states in current-carrying Bi\(_2\)Se\(_3\) samples. This is evident from the top curves in Fig. 4(a)–(d).

Furthermore, it is clear from the Fig. 5 that the asymmetry in tunneling currents is increased with the increase in tin doping levels, from about 5% for the intrinsic sample to 21% for sample C (for the case of negative \( V_{gap} \)). This can be explained as follows. Tin doping increases the bulk resistance, but does not affect the physical properties of the conducting surface mode because it is topologically protected. This leads to the increase in the surface portions of bias currents through Bi\(_2\)Se\(_3\) samples with higher doping and, consequently, results in the observed increase in tunneling current asymmetry.

Special efforts were made to ensure that the obtained results clearly reveal the accumulation of spin-polarized electrons on the surface of the current-carrying samples and were not contaminated by possible thermal instability of Fe-coated tips. This was achieved by repeating our experiments at room temperature with the same tip and the same samples in a span of several days and obtaining the same results.

It was also found that the described asymmetry in tunneling currents is enhanced for smaller values of these currents. This is illustrated in Fig. 5. Asymmetry as a function of tin doping measured at \( I_t^- = 100 \text{ pA} \) and \( V_{gap} = \pm 50 \text{ mV} \).
In this letter, a novel experimental technique for the STM detection of spin-polarized electron accumulation on the surface of topological insulators is presented. The observed changes (asymmetries) in the values of the tunneling currents upon reversal of the direction of bias currents through Bi$_2$Se$_3$ samples are clear experimental signatures of surface spin-polarized electron accumulations. Furthermore, the observed increase in the aforementioned asymmetry with the increase in tin doping levels of Bi$_2$Se$_3$ samples, reveals the suppression of bulk conductivity and consequent enhancement of surface spin-polarized electron accumulations in Bi$_2$Se$_3$.

The presented results suggest that the STM-based measurements with iron-coated tungsten tips may open new opportunities in the study of surface effects in topological insulators. These measurements are very local in nature (i.e., they are on the nanoscale). Hence, the described technique can be extended to study the correlation of these effects with surface morphology of topological insulators.

III. CONCLUSION

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