Topological insulator based spin valve devices: Evidence for spin polarized transport of spin-momentum-locked topological surface states

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A. Topological insulators
B. Spin valve
C. Spin-momentum locking
D. Spin polarized current

1. Introduction

Three-dimensional (3D) topological insulators (TIs) represent an interesting new class of quantum matter hosting spin helical surface states protected by time-reversal symmetry [1–7]. The nontrivial topological surface states (TSS, depicted in Fig. 1a) located inside the bulk band gap feature a characteristic spin-momentum-locking, where charge carriers of a given momentum (k) are spin polarized in-plane perpendicularly "locked" to \( \vec{k} \). For electrons, the spin polarization (\( \vec{S} \)) is along the direction of \( \vec{k} \times \vec{n} \) (\( \sigma^- \) helicity, governed by the left hand rule, depicted in Fig. 1b) with \( \vec{n} \) being the surface normal, and holes have the opposite polarization (\( -\vec{k} \times \vec{n} \), \( \sigma^+ \) helicity, right handed spin-momentum locking). A directional electrical current (I) carried by such spin-helical TSS would be automatically spin-polarized (noting the spin polarization for a given current direction is the same regardless whether the current is carried by electrons or holes, as electron momentum is opposite to the current direction), and its spin polarization reverses upon reversing the current direction (depicted in Fig. 1c,e), or going to the opposite surface (reversing \( \vec{k} \)). The spin-momentum locking of TSS is the basis of the topological protection (as a backscattering that reverses momentum would have to reverse the spin) and many other exotic physics predicted for TI (e.g. majorana fermions) [4,8], and the expected helical spin-polarized transport makes TI particularly promising for spintronics device applications [4,9–13]. While the existence of the spin-momentum-locked TSS in 3D TIs has been established by spin and angle resolved photoemission spectroscopy (spin ARPES) measurements [14–20], direct demonstration of the spin-helical current (current induced spin polarization) using spin-sensitive transport measurements has been lacking till very recently [21–23], even though various different theoretical proposals have been discussed [9–13]. Previously, the spin valve effect (where a current flows through two ferromagnets (FM) of parallel magnetizations with lower resistance and antiparallel magnetizations with higher resistance) and spin valve devices have been commonly used to study spin transport in various materials.
The high quality bulk Bi$_2$Se$_3$ single crystal is grown by the Bridge- man method [31–33]. Thin flakes of 10–20 nm in thickness are exfoliated from the bulk crystal using the standard “scotch tape” method [31,34–35] and are placed on top of heavily doped Si substrates with 300 nm SiO$_2$. The FM electrodes (Ni, thickness=40 nm, length ~3 μm, width between 200 nm and 800 nm) crossing and contacting the TI top surface are defined by standard e-beam lithography and deposited by e-beam evaporation. These Ni electrodes are contacted further outside the Bi$_2$Se$_3$ flake by Au electrodes fabricated by a second e-beam lithography and evaporation. In this work, we have selected flakes of relatively narrow width (~1 μm) and performed two-terminal spin-valve measurements (resistance between two FM electrodes) using a DC bias current $I$ and an in-plane $B$ field (see Fig. 1c–f for device and measurement schematics). The voltage ($V$) difference is measured between the FM (Ni) electrodes (labeled by Ni1 and Ni2), and the magnetoresistance (MR) is defined by $R = |V/I|$. Hereafter, we define $+I$ (−$I$) direction as from Ni1 to Ni2 (Ni2 to Ni1) along +$x$ (−$x$) axis and the positive (negative) in-plane $B$ field points to the +$y$ (−$y$) axis indicated by the green and yellow arrows, respectively, as depicted in Fig. 1c. At a fixed bias current $I$, we sweep the $B$ field from a sufficiently large positive value (far exceeding the coercive fields of the Ni electrodes, so that both Ni electrodes are magnetized along +$y$ direction, depicted in Fig. 1d) through zero and to a large negative value (both Ni electrodes magnetized along −$y$ direction, depicted in Fig. 1c) and then sweep back again to the starting large positive $B$ field. We then reverse the direction of the bias current and repeat the above measurements (Fig. 1e,f). Results from two devices are presented below.

2. Experimental

The high quality bulk Bi$_2$Se$_3$ single crystal is grown by the Bridge- man method [31–33]. Thin flakes of 10–20 nm in thickness are exfoliated from the bulk crystal using the standard “scotch tape” method [31,34–35] and are placed on top of heavily doped Si substrates with 300 nm SiO$_2$. The FM electrodes (Ni, thickness=40 nm, length ~3 μm, width between 200 nm and 800 nm) crossing and contacting the TI top surface are defined by standard e-beam lithography and deposited by e-beam evaporation. These Ni electrodes are contacted further outside the Bi$_2$Se$_3$ flake by Au electrodes fabricated by a second e-beam lithography and evaporation. In this work, we have selected flakes of relatively narrow width (~1 μm) and performed two-terminal spin-valve measurements (resistance between two FM electrodes) using a DC bias current $I$ and an in-plane $B$ field (see Fig. 1c–f for device and measurement schematics). The voltage ($V$) difference is measured between the FM (Ni) electrodes (labeled by Ni1 and Ni2), and the magnetoresistance (MR) is defined by $R = |V/I|$. Hereafter, we define $+I$ (−$I$) direction as from Ni1 to Ni2 (Ni2 to Ni1) along +$x$ (−$x$) axis and the positive (negative) in-plane $B$ field points to the +$y$ (−$y$) axis indicated by the green and yellow arrows, respectively, as depicted in Fig. 1c. At a fixed bias current $I$, we sweep the $B$ field from a sufficiently large positive value (far exceeding the coercive fields of the Ni electrodes, so that both Ni electrodes are magnetized along +$y$ direction, depicted in Fig. 1d) through zero and to a large negative value (both Ni electrodes magnetized along −$y$ direction, depicted in Fig. 1c) and then sweep back again to the starting large positive $B$ field. We then reverse the direction of the bias current and repeat the above measurements (Fig. 1e,f). Results from two devices are presented below.

3. Results and discussions

Fig. 2 shows the results of magnetoresistance measurements in our spin valve device “A”, fabricated on a 12 nm-thick exfoliated
Bi$_2$Se$_3$ flake. The inset of Fig. 2a shows the optical image of the device, where the spacing between the two Ni electrodes is 200 nm. The measurements were made using bias current $I = 100$ nA and at temperature $T = 0.3$ K. The relatively high 2-terminal resistance ($\sim 200$ k$\Omega$) of this device is attributed to a large contact resistance likely resulted from an unclean interface between TI and contacts (e.g. due to surface contaminants from fabrication process). The MR measured between $-2$ T and $+2$ T is shown in Fig. 2a and b and a zoomed-in view (between $-0.5$ T and $+0.5$ T) shown in Fig. 2c and d. The first set of features one may notice are some drastic resistance jumps observed at very low B field ($<0.4$ T), where $R$ vs $B$ is hysteretic and goes up and down several times. Such complicated features at low $B$ are quite different from the MR features observed in previously studied planar spin-valve devices on non-TI materials (where the MR typically displays a single resistance “bump” on each side of zero $B$ field depending on the $B$ field sweep direction) [24–28], and most of these features we observed remain to be better understood. They may be related to the spin valve effects between the two Ni electrodes (whose magnetization switches at such low fields) that may also involve the polarized spin of TSS in the TI channel in a way that substantially modifies the usual spin-valve behavior seen in non-TI materials. Furthermore, the formation and switching of multiple different magnetic domains in the Ni electrodes could also play some roles. However, most of these complicated features at low $B$ are not always observed in other devices we studied (see Fig. 3), and are not the focus of this paper. Our main feature in the MR that we focus on here is the more subtle asymmetry in the MR between large positive ($B > 0.5$ T) and large negative ($B < -0.5$ T) fields, where the magnetization $\vec{M}$ in both the FM electrodes are parallel and points along $+y$ or $-y$ directions for such large $+B$ or $-B$ field respectively. In Fig. 2a, the asymmetric MR manifests as a “high $R$” state of $\sim 196$ k$\Omega$ for $B < -0.4$ T and a “low $R$” state of $\sim 193$ k$\Omega$ for $B > 0.4$ T, observed in the MR data from both $B$ field sweep directions. Such an unusual asymmetry in MR between opposite large $B$ fields is not observed in previously studied spin valve devices in non-TI materials (even spin–orbit coupled semiconductors such as InAs [25]), where the “asymmetric” MR at large $B$ fields (when the two FM electrode share the same magnetization direction) is the same between the opposite $B$ field directions (when the two FM electrodes both reverse their common magnetization direction) [24–28]. The asymmetry in MR could be consistent with the existence of a substantial spin polarization ($\vec{S}$) in the channel that is not reversed when the $B$ field is reversed. Most strikingly, we find that the “polarity” of the above asymmetry in MR is reversed by reversing the current direction ($I \sim -100$ nA, as shown in Fig. 2b,d), where the “high $R$” state now occurs for $B > 0.4$ T and “low $R$” state now occurs for $B < -0.4$ T, suggesting that channel spin polarization is reversed by reversing $I$ (thus “locked” to the current direction). We can further define a normalized spin-valve signal $\beta$ in terms of the asymmetry (difference between large $+B$ and $-B$) in relative MR $\Delta R/R_0 = (R(B) - R_0)/R_0$, $R_0$ being the average of the $R(B)$, as shown in the right axes of Fig. 2. For device “A”, we find $\beta$ is around 1–2%.

We have also performed similar spin valve measurements in another device “B” and further studied the temperature effect, shown in Fig. 3. Device “B” is fabricated from a 20 nm-thick exfoliated Bi$_2$Se$_3$ flake and our measurements were performed with bias $I = \pm 1$ $\mu$A at $T = 1.4$ K and 10 K. At $T = 1.4$ K, shown in Fig. 3a,b, we see again the asymmetric MR between large $+B$ and $-B$ fields (with also a clear hysteresis near zero field), consistent with the spin valve effect between TSS and FM electrodes (with a normalized spin valve signal $\beta \sim 0.1\%$, much lower than that of device “A”). We note that the more complicated features at low $B$ (in Fig. 2 for device “A”) are not observed here in device “B” within experimental resolution. As $T$ was increased to 10 K, such
asymmetric in the MR is no longer observable (Fig. 3c,d) within the experimental resolution. This weakening and disappearance of spin-valve signal at elevated $T$ may be related to increased scattering of carriers, or thermal activation of carriers from spin polarized TSS to bulk conduction bands (which may even carry spin-valve signal at elevated temperature). This weakening and disappearance of asymmetric in the MR is no longer observable (Fig. 3c,d) within the experimental resolution.

Our observation can be qualitatively understood as a spin-valve effect between the current-induced spin polarization of TSS on the TI top surface and the spin-polarized FM contacts as depicted in Fig. 1c–f. Here, we focus on the case under a large in-plane $B$ field such that the magnetization ($\vec{M}$) of the FM electrodes has the same orientation (either $+\gamma$ or $-\gamma$) along the easy axis. Inspired by the well-known spin valve and GMR effect between two ferromagnets with antiparallel orientation (leading to high $R$) or parallel (leading to low $R$) [24–28,37,38], one expects a high $R$ state when the spin polarization $\vec{S}$ (whose direction is determined by the current direction according to the spin-momentum locking of TSS, as depicted in Fig. 1) in the channel (top surface of TI) is antiparallel (“in disagreement”) with the orientation of $\vec{M}$ (determined by the direction of $B$ field) in both FM electrodes (Fig. 1d,e), while a low $R$ state when $\vec{S}$ and $\vec{M}$ are parallel (“in agreement”, Fig. 1c,f). Reversing the direction of (large) $B$ field reverses $\vec{M}$ but does not change $\vec{S}$, thus giving rise to an asymmetry in MR between large $+B$ and $-B$ fields. Reversing $I$ reverses $\vec{S}$, thus reversing the “polarity” of the MR asymmetry. Our model suggests that the two-terminal MR of a TI-FM spin valve device is not only controlled by the magnetization of the FM electrodes, but also the applied DC bias $I$. This model further suggests that there would be a “symmetry” (expecting the same MR) if both $B$ field ($\vec{M}$) and current $I$ ($\vec{S}$) are simultaneously reversed. We note that such a symmetry (upon reversing both $B$ and $I$) does indeed hold approximately for the data shown in Fig. 2 (the MR curve measured sweeping from $+B$ to $-B$ at $I$ nearly reproduces the curve measured sweeping from $-B$ to $+B$ at $-I$).

Recently and during the preparation of this paper, we became aware of the work by Li et al. [21] which reports a transport signature of spin-momentum-locking of TSS in MBE (molecular beam epitaxy)-grown Bi$_2$Se$_3$ devices (of much larger size than ours). Their measurements appear to be in the linear response regime (with a voltage signal that is proportional to the current $I$, thus a current-independent resistance, and the voltage reverses under $B$ reversal). In contrast, our measurement is in the non-linear response regime (with a resistance signal that depends on the current $I$ and $B$ directions), and cannot be described by the Onsager relationship (which states two-terminal resistance should be symmetric with $B$ field) that only applies to linear-response regime. Our observed asymmetry in MR thus provides another signature of spin-helical transport in TSS. In addition, we also noted 2 other preprints reporting signatures of spin-helical TSS transport measured using spin torque [23] and spin pumping [22] techniques.

4. Conclusions

We have fabricated spin valve devices on exfoliated Bi$_2$Se$_3$ thin films and performed two-terminal spin valve (magneto-resistance, with in-plane $B$ field) between two FM contact electrodes (magnetized by the $B$ field). By driving a DC current, we find that the two-terminal resistance is asymmetric between large positive and negative $B$ fields. The “polarity” of the asymmetry can be reversed by reversing the direction of the bias current. Furthermore, the measured resistance asymmetry decreases as temperature increases. Our observation is consistent with the spin-momentum helical locking of TSS producing a spin-polarized helical current, and opens ways to utilize such a remarkable property of TI for future applications in nanoelectronics and spintronics.

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