



<b>Title</b>	<b>Interplay between topological insulators and superconductors</b>
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## Interplay between topological insulators and superconductors

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Topological insulators are insulating in the bulk but possess metallic surface states protected by time-reversal symmetry. Here, we report on a detailed electronic transport study in high-quality  $\text{Bi}_2\text{Se}_3$  topological insulator thin films contacted by superconducting (In, Al, and W) electrodes. The resistance of the film shows an abrupt and significant upturn when the electrodes become superconducting. In turn, the  $\text{Bi}_2\text{Se}_3$  film greatly weakens the superconductivity of the electrodes, significantly reducing both their transition temperatures and their critical fields. A possible interpretation of these results is that the superconducting electrodes are accessing the surface states and the experimental results are consequences of the interplay between the Cooper pairs of the electrodes and the spin-polarized current of the surface states in  $\text{Bi}_2\text{Se}_3$ .

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### I. INTRODUCTION

Bismuth-based materials have long been studied for their thermoelectric properties.<sup>1-3</sup> Bismuth selenide ( $\text{Bi}_2\text{Se}_3$ ), bismuth antimonide ( $\text{Bi}_{1-x}\text{Sb}_x$ ), bismuth telluride ( $\text{Bi}_2\text{Te}_3$ ), and antimony telluride ( $\text{Sb}_2\text{Te}_3$ ) have been predicted theoretically and confirmed experimentally by angle-resolved photoemission spectroscopy (ARPES) experiments to be three-dimensional (3D) topological insulators (TIs) because of strong spin-orbit interactions.<sup>1-11</sup> In transport measurements of 3D TIs,<sup>12-19</sup> quantum magnetoresistance (MR) oscillations have been observed and interpreted as evidence of a topologically protected surface state. Of these,  $\text{Bi}_2\text{Se}_3$ , with a simple surface state structure (a single Dirac cone) and relatively large band gap (0.3 eV) has become a reference material in 3D TIs. A key feature of the surface state is that the spin and momentum of the conduction electrons are locked,<sup>4</sup> which has been confirmed by ARPES measurements<sup>20,21</sup> but has not been directly demonstrated in transport experiments.

In this paper, we report on the transport behavior of crystalline  $\text{Bi}_2\text{Se}_3$  films contacted by three kinds of superconducting electrodes to study the interplay between the superconductivity and the TI surface state. We use superconducting bulk indium (In) electrodes and mesoscopic aluminum (Al) and tungsten (W) electrodes to study the transport property of the  $\text{Bi}_2\text{Se}_3$  films with thicknesses of 5 and 200 quintuple layers (QLs) on sapphire and silicon substrates, respectively. Every QL is 1 nm thick. A simple two-probe configuration is used to minimize the fabrication processing of the electrodes and hence to reduce the risk of altering the intrinsic property of the TI samples. The two-probe (pseudo-four-probe) geometry used here has two contact pads (wires) for each probe. The distances between superconducting electrodes are 1 mm (In) and 1  $\mu\text{m}$  (Al and W). Irrespective of the material of the electrodes, the thickness of the  $\text{Bi}_2\text{Se}_3$  film, the separation

of electrodes, the substrates, and the contact resistance, the low-bias resistance shows a large and abrupt increase near the superconducting transition temperature ( $T_C$ ) of the electrodes. Most interestingly, we observe that the  $\text{Bi}_2\text{Se}_3$  films reduce both the  $T_C$  and the critical field ( $H_C$ ) of the superconducting electrodes significantly.

### II. EXPERIMENTAL RESULTS

Recent progress in thin film growth of TIs by molecular beam epitaxy (MBE) has made planar TI devices possible.<sup>22-26</sup> Our high-quality  $\text{Bi}_2\text{Se}_3$  films were grown under Se-rich conditions on sapphire (5 QL) and high-resistivity silicon (200 QL) substrates in ultrahigh-vacuum MBE systems. A scanning tunneling microscope (STM) image of a 5-QL sample is shown in the left inset of Fig. 1(a). The atomically flat morphology demonstrates the high crystal quality of the film. The carrier density and the mobility of the 5-QL film at 2 K are  $\sim 4 \times 10^{18} \text{ cm}^{-3}$  ( $2 \times 10^{12} \text{ cm}^{-2}$ ) and  $3320 \text{ cm}^2/\text{Vs}$  by Hall measurement, respectively. With decreasing thickness of the film, the surface-to-volume ratio increases and surface properties should become more prominent. However, it has been shown by ARPES that in films with thickness of less than 5 QL, the interaction between top and bottom surfaces may destroy the topologically protected surface state.<sup>23</sup>

The right inset of Fig. 1(a) is a schematic diagram of our transport measurement structure. Superconducting In dots  $\sim 0.5 \text{ mm}$  in diameter and  $\sim 0.2 \text{ mm}$  thick are directly pressed onto the top surface of the  $\text{Bi}_2\text{Se}_3$  film. The distance between the two electrodes is  $\sim 1 \text{ mm}$ . Figure 1(a) shows resistance as a function of temperature ( $R-T$ ) for the 5-QL  $\text{Bi}_2\text{Se}_3$  film. In this paper, unless noted otherwise, the magnetic field is always applied perpendicular to the film and the excitation current for the measurement is 50 nA (corresponding essentially to a zero-bias resistance measurement). From 300 to 45 K, the  $R-T$

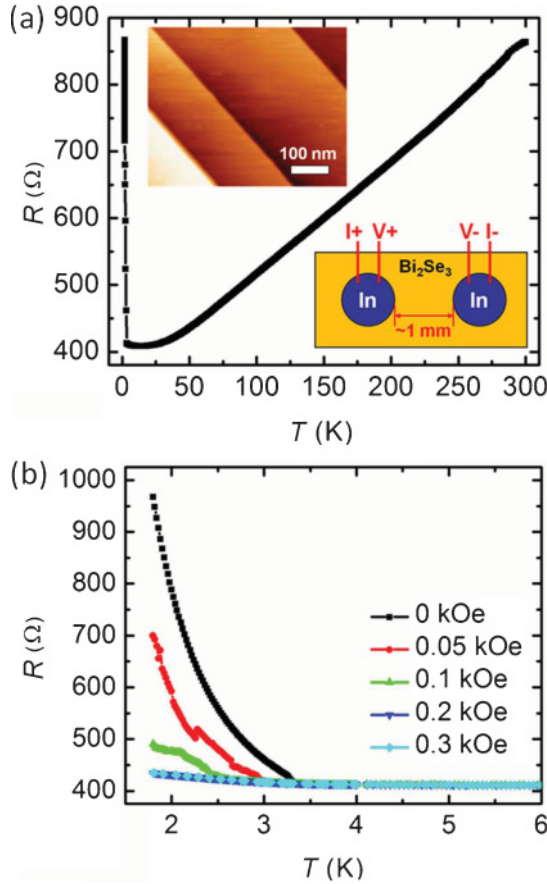


FIG. 1. (Color online)  $R$ - $T$  behavior of the 5-nm-thick  $\text{Bi}_2\text{Se}_3$  film contacted by two superconducting In dots. (a)  $R$  vs  $T$  of the 5-QL  $\text{Bi}_2\text{Se}_3$  film from room temperature to low temperature. The left inset is a STM image of the  $\text{Bi}_2\text{Se}_3$  film. The right inset is the measurement structure. (b)  $R$  vs  $T$  at different perpendicular fields. The curves at 0.2 and 0.3 kOe are superimposed.

curve shows linear metallic behavior. A resistance minimum is found near 13.3 K. The residual resistance ratio between 300 and 13.3 K is 2.1. Below 13.3 K, the resistance increases gradually with decreasing temperature. However, at 3.29 K (slightly below the  $T_C$  of bulk In, or 3.4 K), the resistance shows an abrupt increase. This resistance enhancement is shown in more detail in Fig. 1(b). The resistance at 1.8 K (967.23  $\Omega$ ) is 2.34 times the resistance when the In electrodes are normal at  $T = 3.4$  K. With an increasing field, this resistance enhancement decreases rapidly. When the field is 200 Oe, the enhancement behavior is suppressed. This means the actual critical field of the In electrodes here is lower than 200 Oe, which is the critical field of bulk In at 1.8 K. We interpret the enhancement in  $R$  to be a consequence of the onset of superconductivity of the In electrodes; however, it appears that the transition temperature and critical field of the In electrodes when contacting the  $\text{Bi}_2\text{Se}_3$  film are slightly below the natural values.

Resistance as a function of the magnetic field ( $R$ - $H$ ) for the 5-QL  $\text{Bi}_2\text{Se}_3$  film is shown in Fig. 2(a). At 4 K (above  $T_C$  of In), the  $R$ - $H$  curve shows linear MR from 26 to 80 kOe. Such a linear MR has been attributed to the surface states with the linear energy-momentum correlation.<sup>26,27</sup> However, at 1.8 K

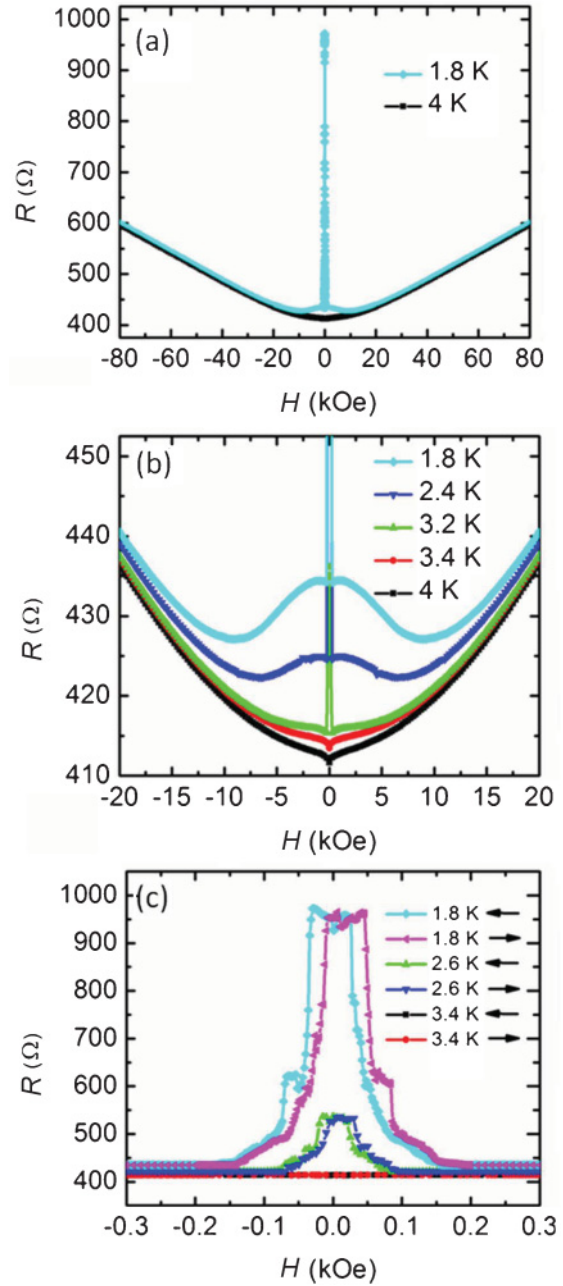


FIG. 2. (Color online)  $R$ - $H$  scans of the 5-nm-thick  $\text{Bi}_2\text{Se}_3$  film contacted by In electrodes. (a) Resistance as a function of perpendicular magnetic field at 4 and 1.8 K. (b) Magnified MR for several temperatures. (c) MR peaks near the zero field show terrace structure and hysteresis for scans made below  $T_C$ . The  $\rightarrow$  arrow indicates the scan was made from a negative to a positive field.

(below  $T_C$  of In), near the zero field, the sample exhibits a striking MR peak. Between 0.2 and 9 kOe, well above the critical field of the electrodes, the resistance decreases unexpectedly with the field. Upon further increase in the magnetic field, the MR shows the same positive linear behavior as the  $R$ - $H$  curve at 4 K.

Figure 2(b) shows MR in a small field at different temperatures in more detail. Above  $T_C$  of the In electrodes (at 3.4 and 4.0 K), we observe a positive MR. In addition, there is a small MR dip around the zero field, which has been studied

carefully and attributed to the weak antilocalization effects in TIs.<sup>26,28</sup> At temperatures below  $T_C$  of the In electrodes, the MR dip disappears and a sharp MR peak emerges. With decreasing temperature, the peak value increases rapidly, consistent with the  $R$ - $T$  curves of Fig. 1. At 1.8 and 2.4 K, besides the sharp resistance peak around the zero field, an additional negative MR is observed from 200 Oe to 9 and 7 kOe, respectively. At lower temperatures, the negative MR is more robust. This result is unexpected. TI films contacted with normal metal electrodes show positive MR in a perpendicular field;<sup>28</sup> therefore, the negative MR cannot be from the TI film itself. However, if the observed negative MR is due to the superconductivity of In electrodes, we would not expect this behavior for fields larger than the  $H_C$  of the In electrodes ( $\sim 200$  Oe at 1.8 K). Interestingly, this negative MR behavior extends up 9 kOe—but only at temperatures below  $T_C$  of the electrodes.

Figure 2(c) shows the details of the MR peak shown in Figs. 2(a) and 2(b). Under higher field resolution, the MR “peak” appears as a plateau with terraces. When we scan magnetic field from negative to positive values and then from positive back to negative values, the sample exhibits hysteretic behavior at 2.6 and 1.8 K. The plateau/terrace structure of the MR peak and the hysteresis are suggestive of a ferromagnetic response in the conduction electrons. There is no possibility of magnetic contamination in the process of sample preparation. A 3D image of the resistance as a function of field and temperature and the resistance contour map along the  $T$ - $H$  axes, constructed from the experimental data we have obtained on this sample, are shown in Fig. 3. More details can be revealed in this figure. In addition to bulk In electrodes measurements, mesoscopic superconducting Al and W electrodes were patterned on the TI films to test the universality of the observed phenomena. The inset of Fig. 4(a) is a scanning electron microscope (SEM) image of our measurement structure. The  $\text{Bi}_2\text{Se}_3$  film for this sample is 200 nm thick and grown on high-resistivity silicon substrate. The substrate is completely insulating below 150 K. The carrier density and the mobility of the film at 1.8 K are found to be  $2.76 \times 10^{18} \text{ cm}^{-3}$  ( $5.52 \times 10^{13} \text{ cm}^{-2}$ ) and  $2800 \text{ cm}^2/\text{Vs}$  by Hall measurement, respectively. The superconducting Al electrodes are 50 nm thick and directly deposited on the top surface of the film by electron beam lithography followed by electron-beam-assisted evaporation. The distance between the two Al electrodes is  $1 \mu\text{m}$ . Figure 4(a) shows the  $R$ - $T$  curves of this sample. Under a zero magnetic field, there is a sharp resistance increase at 0.95 K from  $23.5 \Omega$ , which becomes saturated below 0.85 K at  $28.5 \Omega$ . This enhancement is similar to our observation in the sample with bulk In electrodes. The onset temperature of the resistance enhancement at 0.95 K is significantly lower than the  $T_C$  of 50-nm-thick Al film. This enhancement is suppressed by a field of 100 Oe. An Al film of the same thickness that was evaporated with the same procedures on an insulating  $\text{Si}_3\text{N}_4$  substrate shows a  $T_C$  of 1.4 K and a critical field of  $\sim 800$  Oe at 0.65 K. Thus, the superconductivity of the Al electrodes is substantially and clearly weakened by the TI film. As shown previously, the effect of the  $\text{Bi}_2\text{Se}_3$  film on the bulk In electrodes is not as strong. This is not unreasonable, because the Al electrodes are only 50 nm thick and the In electrodes are “macroscopic” in size. The MR behavior in a small field, as shown in Fig. 4(b), is

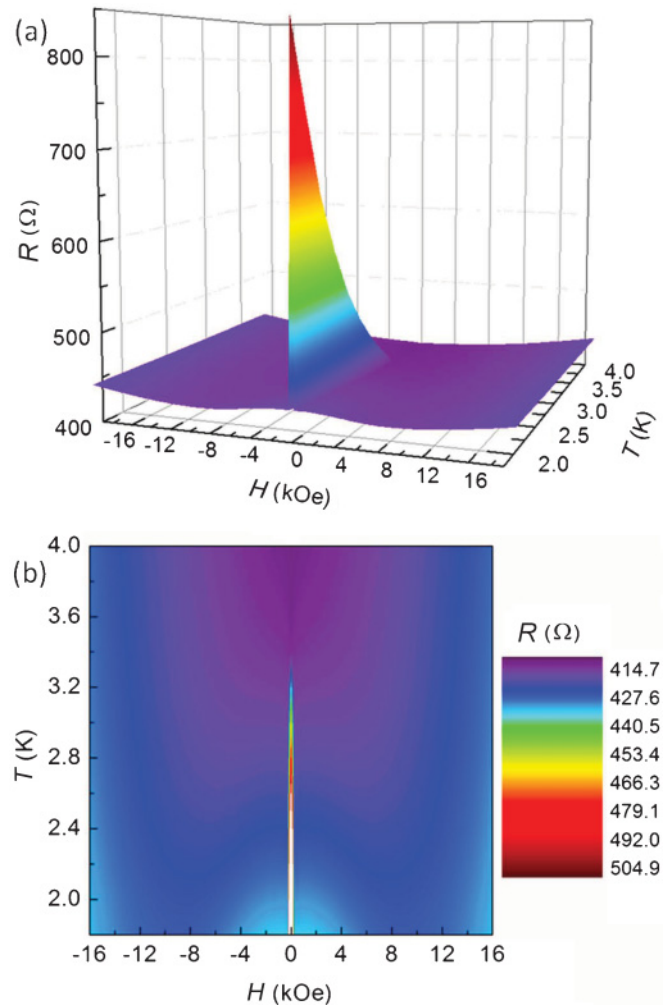


FIG. 3. (Color online) (a) 3D image of resistance as a function of the perpendicular field and temperature measured with a bias current of 500 nA for the 5-QL-thick  $\text{Bi}_2\text{Se}_3$  film contacted by bulk In electrodes (In- $\text{Bi}_2\text{Se}_3$ -In). A sharp resistance enhancement induced by the interaction between superconducting electrodes and  $\text{Bi}_2\text{Se}_3$  film is found. (b) Color contour map of resistance along the temperature and perpendicular magnetic field axes of the 5-nm-thick  $\text{Bi}_2\text{Se}_3$  film contacted by In electrodes. The colors represent resistance from  $410 \Omega$  (deep purple, the shade at the top of the color scale) to  $510 \Omega$  (deep red, the shade at the bottom of the color scale). White means the resistance is larger than  $510 \Omega$ .

similar to that found with In electrodes, as shown in Fig. 2(c). The observations in this Al electrode device were confirmed in two additional devices with the same geometry by a zero-bias differential resistance measurement, which was carried out using a lock-in amplifier with 100-nA alternating current excitation at a frequency of 97 Hz.

By means of the focused ion beam (FIB) deposition technique,<sup>29–32</sup> superconducting W electrodes were fabricated on the  $\text{Bi}_2\text{Se}_3$  film [inset of Fig. 5(a)]. The thickness of the  $\text{Bi}_2\text{Se}_3$  sample is also 200 nm, and the distance between two W electrodes is  $1 \mu\text{m}$ . The FIB-deposited amorphous W strips have been used in a number of experiments as superconducting electrodes.<sup>29–31</sup> The  $T_C$  of the strips depends on the exact deposition parameters of the FIB process, but

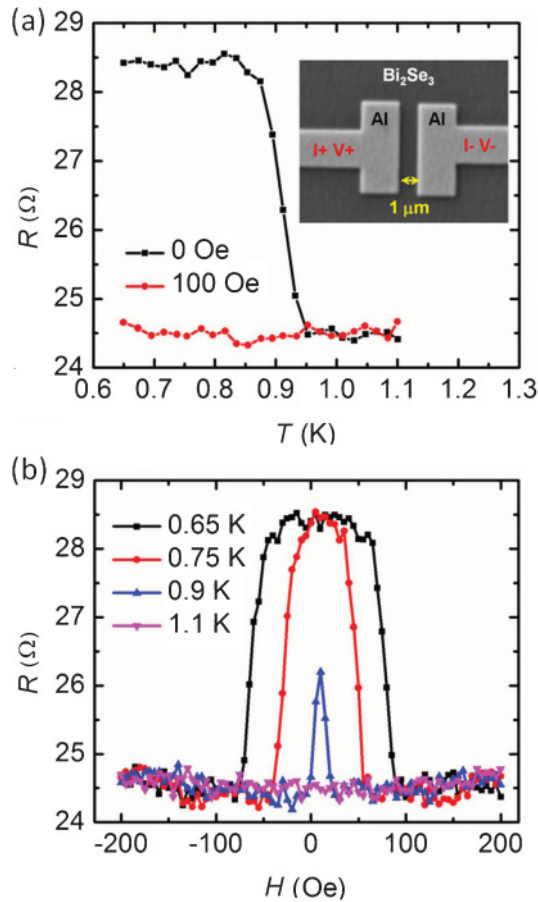


FIG. 4. (Color online) Transport behaviors of 200-nm-thick  $\text{Bi}_2\text{Se}_3$  films contacted by superconducting Al electrodes. (a) A sharp resistance enhancement is seen at 0.95 K, which saturates below 0.85 K. An applied magnetic field of 100 Oe suppresses the enhancement. The inset is a SEM image of the Al contacts on the surface of the  $\text{Bi}_2\text{Se}_3$  film. (b) The details of the MR in a small field. The resistance peak at 0.65 K is suppressed under a field of less than 100 Oe, which is much smaller than the  $H_C$  (800 Oe) of a 50-nm-thick Al film not contacting a  $\text{Bi}_2\text{Se}_3$  film. The  $T_C$  of such an “isolated” Al film is 1.4 K.

it was consistently found to be between 4 and 5 K when contacting metallic and magnetic nanowires.<sup>29–32</sup> This is much higher than the  $T_C$  of pure W ( $\sim 12$  mK), because the FIB-deposited W strips contain approximately 40% atomic carbon and 20% atomic gallium.<sup>32</sup> During the FIB deposition process, the top layers of the  $\text{Bi}_2\text{Se}_3$  film are etched away, making fresh contact between the electrodes and the film. It has been shown that in the FIB process, electrically transparent interfaces were achieved.<sup>30</sup> Figure 5(a) shows the  $R$ - $T$  curves of the W- $\text{Bi}_2\text{Se}_3$ -W structure at different fields. The superconductivity-induced resistance enhancement is again seen in this structure. The resistance increases from 0.5 to 6.5  $\Omega$  when the W electrodes turned superconducting. The onset temperature of the resistance enhancement is  $\sim 3.5$  K, which is again significantly smaller than the  $T_C$  (4–5 K) of the W strips.<sup>29–31</sup> The magnetic field sufficient to suppress the resistance peak is less than 10 kOe at 2.2 K, which is again much smaller than the  $H_C$  of the W strips ( $\sim 80$  kOe).<sup>29,30</sup> These results confirm the findings with In and Al electrodes

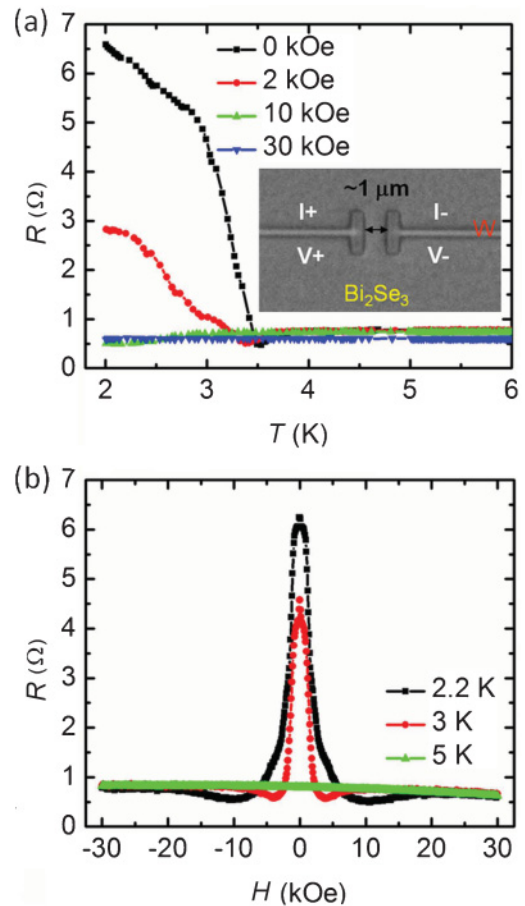


FIG. 5. (Color online) Transport behaviors of a 200-nm-thick  $\text{Bi}_2\text{Se}_3$  film contacted by superconducting W electrodes. (a)  $R$  vs  $T$  scans under different magnetic fields. The inset is a SEM image of the W contacts on the surface of the  $\text{Bi}_2\text{Se}_3$  film. (b) MR at different temperatures. When the W electrodes become superconducting, the MR shows a large peak around the zero field. This behavior disappears when the temperature is larger than the  $T_C$ . The resistance peak is suppressed under a field of  $\sim 10$  kOe at 2.2 K, much smaller than the  $H_C$  of the W strips (80 kOe).

that the  $\text{Bi}_2\text{Se}_3$  film weakens the superconductivity of the contacting electrodes. The MR behavior shown in Fig. 5(b) is also consistent with that shown in Fig. 2. For MR scans made below  $T_C$  of W, in addition to the prominent peak at low field, a minimum in  $R$  is found at a field value above  $H_C$  of the specific temperature of the scan.

To further understand the interplay among the TI thin films with superconducting electrodes, we carried out differential conductance measurements. Figure 6(a) plots field-dependent  $I$ - $V$  characteristics of the W- $\text{Bi}_2\text{Se}_3$ -W sample at 0.5 K. There is a sudden voltage drop (negative conductance) when the field is less than 1.5 kOe in  $I$ - $V$  curves. This phenomenon is not fully understood. Figure 6(b) shows bias-dependent differential conductance ( $dI/dV$ ) of the same sample at different fields at 0.5 K. The negative  $dI/dV$  for small field ( $H < 2$  kOe) is due to the voltage drop in Fig. 6(a). Apart from its negative value, the differential conductance in the zero field is also strongly suppressed below 0.33 mV ( $\sim \Delta/2e$  of W, where  $\Delta$  is the energy gap of the FIB-deposited W). This differential conductance suppression becomes weaker

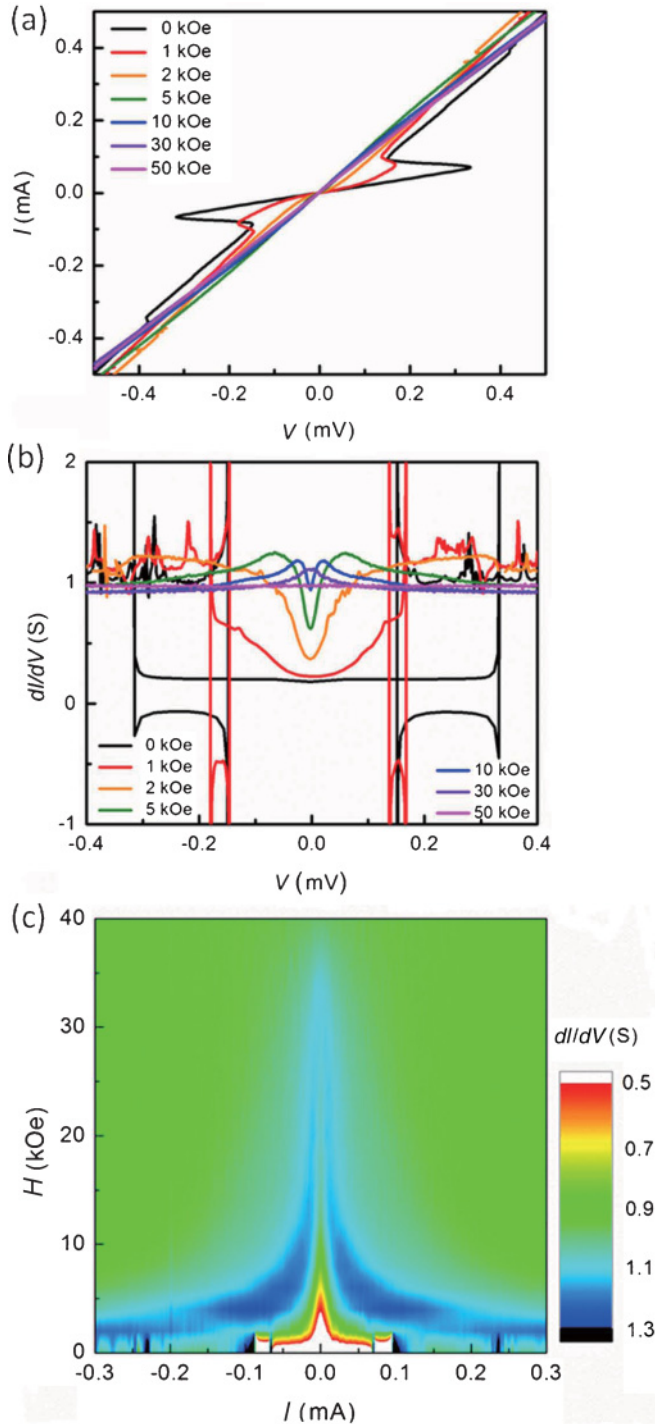


FIG. 6. (Color online) (a)  $I$  vs  $V$  at different perpendicular magnetic fields for the W-Bi<sub>2</sub>Se<sub>3</sub>-W sample at 0.5 K. (b)  $dI/dV$  vs  $V$  at different magnetic fields at 0.5 K. (c)  $dI/dV$  as a function of current  $I$  and field  $H$  at  $T = 0.5$  K.

and moves to a lower bias as the field increases. At 30 kOe, the differential conductance switches from a suppression to an enhancement in a small bias regime. Finally, at 50 kOe, the  $dI/dV$  curve becomes a constant, which means the whole system becomes normal. In Fig. 6(c), we map out the variation of  $dI/dV$  as a function of current and magnetic field at 0.5 K. In a low-field and low-excitation current regime, the

differential conductance is small, which is consistent with the observation of resistance upturn (Fig. 5). At a small current bias, with increasing field, the differential conductance first increases and then decreases. The  $dI/dV$  measurements confirm the observed resistance upturn behavior in such a W-Bi<sub>2</sub>Se<sub>3</sub>-W structure (Fig. 5).

### III. DISCUSSION

The results shown here indicate that the phenomena we have observed are universal and reproducible: they are seen with three kinds of superconducting electrode materials, Bi<sub>2</sub>Se<sub>3</sub> film thicknesses of 5 and 200 nm, and separation of the electrodes of 1 mm and 1  $\mu$ m. The electrodes are attached onto the surface of the Bi<sub>2</sub>Se<sub>3</sub> film by mechanical pressure (In), by electron-beam fabrication (Al) and by the FIB process (W).

If the contact between a superconductor and a normal metal is electrically transparent, the leakage of Cooper pairs into the normal metal can induce superconductivity in the normal metal. Simultaneously, the superconductivity of the superconductor on the other side of the interface can be weakened. This behavior is called the proximity effect.<sup>33</sup> While the observed weakening of the superconductivity of electrodes in our experiment is qualitatively consistent with this effect, the substantial decrease in  $T_C$  and  $H_C$  seen in the Al and W electrodes is unprecedented for superconducting electrodes contacting metallic,<sup>29</sup> ferromagnetic,<sup>30</sup> semiconducting nanowires,<sup>34,35</sup> and a two-dimensional graphene system.<sup>36</sup> The observed increase in  $R$  of the Bi<sub>2</sub>Se<sub>3</sub> film when the electrodes turned superconducting is contrary to the conventional proximity effect interpretation and has not been observed in the half-metallic film employing the same measurement configuration.<sup>37</sup>

Extensive studies of transport across superconductor/semiconductor interfaces have shown a range of interesting behavior depending upon the transparency of the contact. When the interface is resistive, as typically occurs due to the formation of an interfacial Schottky barrier, an increased zero-bias resistance often accompanies the normal–superconducting transition of the electrode.<sup>38</sup> In past studies of various semiconductor–superconductor interfaces, such changes in resistance were readily understood using the Blonder-Tinkham-Klapwijk (BTK) model and extensions thereof,<sup>39</sup> wherein the interface transparency is a key factor in determining the temperature and bias dependence of the transport. Our observations, however, are quite different from those seen in all past measurements of semiconductor–superconductor junctions. First, they are robust against large variations in the transparency of the contacts. The resistances of the Bi<sub>2</sub>Se<sub>3</sub> film we measured with the In, Al, and W electrodes are 425, 23.5, and 0.5  $\Omega$  in the normal state and 880, 28.5, and 6.5  $\Omega$  in the superconducting state, respectively. These values indicate that while the contacts with the In electrodes may be slightly resistive, the Al/Bi<sub>2</sub>Se<sub>3</sub> and particularly the W/Bi<sub>2</sub>Se<sub>3</sub> interfaces are electrically transparent. Despite the differences in the contact resistance of the three electrodes, the observed phenomena are essentially the same. Second, we find that the upturn in resistance can be strikingly large compared to that seen in past studies of semiconductor–superconductor junctions,<sup>38,39</sup> and it shows a behavior contrary to expectations from the BTK model.<sup>40</sup> We are particularly

surprised by our observation of the huge resistance upturn (1300%) for the W electrodes, as well as the strong suppression of differential conductance at low bias, thus unexpectedly showing the largest effect for the highest transparency contacts. Since we cannot explain our observations using the BTK model, which has been rigorously and extensively applied to a variety of semiconductor–superconductor junctions, we propose that the observed phenomenon is connected to the spin–helical surface states of the  $\text{Bi}_2\text{Se}_3$  film; in addition, it may be a consequence of the entanglement of bulk and surface transport, because we cannot exclude the bulk transport channel in our measurements.

#### IV. CONCLUSION

A possible explanation of our observations reported here is that we are accessing the special property of TI surface states. The spins of the TI surface states are predicted to be helical with fixed spin orientation at a given momentum.<sup>41</sup> In our transport measurement configuration, the collective spin polarization of the TI surface state is aligned by the current.<sup>42</sup> When the electrodes become superconducting, the spin–singlet Cooper pairs are not compatible with the spin-polarized electrons on the TI surface. Spin flip processes must take place at the interface when the Cooper pairs leak from the current source electrode to TI and when the spin-polarized electrons flow from TI to the superconducting sink electrode. This process produces a sharp resistance enhancement below  $T_C$ . The spin-polarized current in turn strongly weakens the superconductivity of the superconducting electrodes. Recently, transport measurements with superconducting electrodes were also made on  $\text{Bi}_2\text{Se}_3$  nanoribbons<sup>43</sup> and flakes,<sup>16,44</sup> showing

a proximity effect and a downturn of zero-bias resistance. There are two possible explanations for the different behavior between the observations reported here and those reported with nanoribbons or flakes: First, the “minimally processed” samples used in our study could allow better preservation of the spin–momentum locked surface states. Another possibility is that the measurements of nanoribbons and micron-sized flakes are carried out in a measurement geometry that is clearly different from the one used in the present paper: the former involves sample edges, whereas the latter does not. Whatever the correct explanation for these observations, we believe that a systematic comparative study between these geometries may hold an important clue to the coupling of superconducting states with TI states, which not only exhibits the proximity effect at the TI and superconductor interface<sup>45</sup> but also offers a platform in searching for Majorana fermions.<sup>46,47</sup>

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