Revealing Intrinsic Superconductivity of the Nb/BiSbTe$_2$Se Interface

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Typically, topological superconductivity is reachable via proximity effect by a direct deposition of superconductor (S) on top of a topological insulator (TI) surface. Here, the double critical current in the Josephson junctions based on the topological insulator is observed in the fabricated planar Superconducting Quantum Interference Devicea. By measuring critical currents as a function of temperature and magnetic field, it is shown that the second critical current stems from the intrinsic superconductivity of the S–TI interface, which is supported by the modified Resistively Shunted Junction model and Transmission Electron Microscopy studies. This complex structure of the interface should be taken into account when the technological process involves Ar-plasma cleaning.

1. Introduction

The topology of electronic band structure plays a central role in explaining different physical phenomena in condensed matter systems.[1–3] The non-triviality of the band structure of some materials leads to the formation of new states of matter, such as topological insulators (TIs),[4] Dirac or Weyl semimetals.[5,6] Furthermore, such non-trivial bands have the potential to induce topological superconductivity via the proximity effect.[7–10] Therefore, the interface between topological insulator and superconductor plays an extremely important role in studying topological superconductivity.[11–14] Since the superconducting order parameter is sensitive to microscale structures around electrodes and their fabrication process, it is crucial to evaluate both the transport properties of superconducting junctions and their local structure to establish topological superconductivity.

In this work, we demonstrate the double critical current in the Superconducting Quantum Interference Device (SQUID), fabricated by sputtering of Nb on top of the Fe-doped BiSbTe$_2$Se flake. By analyzing those critical currents as a function of temperature and magnetic field, we show that the second critical current stems from the intrinsic superconductivity of the S/TI interface, which is supported by the Restively Shunted Junction (RSJ) model and Transmission Electron Microscopy (TEM) imaging. Our deep insight into the interface of the Chalcogenid-based TI junction provides a future qualitative analysis of emergent unconventional SC states and an avoidance strategy for achieving a unitary topological superconducting state for further TI josephsons.

2. Results

We studied Fe-doped 3D topological insulator BiSbTe$_2$Se (Fe-BSTS), synthesized by the modified Bridgman method.[15]
The device is showing very pronounced oscillations of the critical current in the external magnetic field perpendicular to the surface of the loop (Figure 1d). The period of these oscillations is consistent with the flux quantum $\Phi_0 = \hbar c/2e$ per loop area.

The distinctive feature of this device, which can be seen in Figure 1d is what we call “second critical current”. When the external magnetic field corresponds to an integer number of flux quantum, there is only one step in the $I-V$ characteristic, which is identified as critical current. Nevertheless, when the external magnetic field is around $\Phi_0/4$, two clear steps in the $I-V$ characteristic can be seen, which are presented in Figure 1e. We determine the first step as $I_{c1}$ and the second step as $I_{c2}$.

The temperature dependence of critical currents $I_{c1}$, $I_{c2}$ and retrapping current $I_{ret}$ at zero magnetic field are shown in Figure 2a by red rectangles, blue circles and black circles, respectively. The hysteresis of the $I-V$ characteristic is clearly seen in Figure 1d and Figure S3 (Supporting Information). The black dashed line is the fit of $I_{c1}(T)$ by the Kulik–Ormelyanchuk model in the clean limit (KO-2).[19] For details of this model and fitting procedure, see Section S4 (Supporting Information) and Ref. [12].

At large magnetic fields, instead of the standard modulation of the critical current by the Fraunhofer-like dependence, a monotonic decrease of the envelope curve of the critical current is observed (see Figure 2b; Figure S4, Supporting Information). It suggests that the magnetic field acts as a pair-breaking mechanism that suppresses the superconductivity in the surface states,[20] making two critical currents split completely, as shown in Figure 2c. This indicates that the second critical current $I_{c2}$ comes from the properties of the single Josephson junction. Moreover, we have fabricated single Josephson junctions, which also demonstrate this feature (see Section S3, Supporting Information).

3. Discussion

Additional features on the $I-V$ characteristic are frequently observed in Josephson junctions based on topological insulators.[21-22] There could be different explanations, such as Fiske steps,[23] analogs of Shapiro steps,[23,24] McMillan–Rowell[25] or Tomasch[26] resonances. We strongly believe that neither of these explanations fits our experimental results. First of all, our Josephson junctions are fairly overdamped and resonances such as Fiske cannot be observed in our system. Moreover, Shapiro-like resonances with the outer resonant system (for example, sample holder) cannot explain the hysteresis of the $I_{c2}$ we observed, as discussed below. Additionally, single Josephson junctions were measured in the same sample holder, and they show different characteristic voltages, excluding the possibility of unwanted resonances in the sample holder. McMillan–Rowell or Tomasch resonances are also not the cases of our setup as they appear in underdamped JJs with geometrical resonances of quasiparticle or superconducting Andreev states.

Generally, a planar Josephson junction can be described as an $SS'NS'S$ structure.[27] Here, $S$ corresponds to Nb, $S'$ is the...
region of TI under the superconducting electrode, and N is the surface state of the TI, as shown in Figure 2d). Another explanation for the second feature on the I–V characteristic is that \( I_{c2} \) is related to the region \( S' \) located between Nb and TI.

We model our SQUID with an extra superconducting layer \( S' \) underneath the Nb contacts (Figure 2d) with a modified RSJ model. Its equivalent circuit is shown in Figure 2e. For bias currents below \( I_{c2} \), a single JJ in the SQUID acts as an SS’NS’S. When the bias current satisfies the condition \(| I_{b} \| / 2 | + | I_{ss} | > I_{c} \) (see Section S5, Supporting Information, for more details), the JJ acts as an SN’NS’. Here, \( I_{b}, I_{ss}, \) and \( I_{ss} \) are the \( S' \)-layer critical current, bias current, and the current circulating in the loop, respectively. The switching from \( SS'NS'S' \) to \( SN'NN'S \) leads to the second voltage step, which can be described as an appearance of \( N' \)-layer resistance \( R' \) in the shunt.

Numerical simulations predict that \( I_{c2} \) depends on magnetic field as \( A + B \cos(\pi \Phi /\Phi_0) \), which nicely fits the experimental data (Figure 2f). Here, \( A \) and \( B \) are the fit parameters. The \( I_{b,c} \) behavior is fitted as an asymmetric SQUID with an asymmetry coefficient \( \alpha = \frac{I_{b} - I_{c}}{I_{b} + I_{c}} = 0.1 \), where \( I_{b} \) and \( I_{c} \) are the critical currents of the left and the right Josephson contacts in the SQUID.

Typically, the retrapping current is different from the critical current in SNS junctions because of the overheating of the N region.\(^{[26]}\) To describe the fact that \( I_{c1} \) is non-hysteretic but \( I_{c2} \) is (see Figures 2a,f, Section S2 Supporting Information), we added Joule heating in the model by thermally coupling \( S' \) interfaces, N region, S electrode, and the TI flake.\(^{[29]}\) Details of this model could be found in Section S5 Supporting Information. When the bias current reaches the retrapping current, the \( N' \)-layer switches to an \( S' \) and \( R' \) drops to 0. The model suggests that \( R' = R_K \), therefore, the produced heat significantly decreases. The excess heat is being transferred to the flake through metallic surface states, thus the Josephson junction region quickly cools down to the bath temperature. As a result, the SQUID does not exhibit the switching, corresponding to \( I_{c1} \) because of \( I_{c2} < I_{c1} \). That is why we observe that \( I_{c2} \) limits \( I_{c1} \) near the integer flux quantum values (bottom curves in Figure 2f).

In order to describe the blurriness of the second critical current around \( \Phi = \Phi_0 / 2 \) (see Figure 1d), we take into account the inhomogeneity of the \( S' \) layer and treat the \( S' \) as a combination of layers with slightly different \( I_{c} \). Indeed, this approach provides a nonzero width of \( I_{c2} \), if \( R_K^c > R_K > R_K^e \) inequality holds (see Section S5, Supporting Information). However, it does not lead to the full evanescence of \( I_{c2} \) at half flux quantum. Another possible explanation of the blurriness effect is the interplay between the SQUID loop and a superconducting loop formed by \( S' \) region. The difference in London penetration depths and geometric inductance of the loops may result in a peculiar behavior of screening currents that pass through the weak link region and are not considered by the RSJ model.
To validate the presence of the \( S' \) region and to study the \( SS' \) and \( S'N \) interfaces, we performed TEM study of the interface between Nb and Fe-BSTS. We cut one of the Josephson junctions, as shown in Figure 3a along the red line. The corresponding cross-section is shown in Figure 3b. Since we have conducted the TEM study after the transport measurements (and, therefore, thermal cycling) we have observed the same strain-induced buckling feature, as in Ref. [30], which is indicated in Figure 3b by a large rectangle. Interestingly, the interface between strained and unstrained regions is very sharp, as shown in Figure 3c. The unstrained region demonstrates a well-ordered structure, while the strained region is so deformed that there is no order seen. Nevertheless, as in Ref. [30], this feature was formed after the warming up and therefore did not affect our results.

There are regions of specific contrast between the TI layers (region 1) and Nb contacts (region 4) (see Figure 3d). One of them looks darker (region 3) and does not have a periodic structure, while the other (region 2), on the contrary, has a brighter contrast and demonstrates local ordering. A redistribution of the intensity in region 2 can be noticed. In the ordered structure of \( Bi_2SbTe_2Se \), within one quintuple layer, two bright (Bi/Sb) and three less bright (Se/Te) points are observed (see Figure 3e and Figure 1a), while in region 2 the intensity of all atomic columns is more or less the same, which should indicate the averaging of the atomic mass of all atomic columns, that is, mixing of atoms in all or almost all crystallographic positions and a change in the symmetry of the structure. Indeed, the Fourier transform (FFT) taken from the ordered TI layers (see Figure 3e, orange inset) and from region 2 (see Figure 3e, yellow inset) are different. The FFT of the disordered region lacks reflections corresponding to long distances of \( 29.7 \) Å, and the observed image can be interpreted as the \([111]\) zone of an I-centered tetragonal cell with parameters \( a = 4.9 \) Å and \( c = 6.2 \) Å.

The mixed EDX maps (see Figure 3f) show that region 2 is enriched in Bi and Sb although antimony is not shown on the mixed EDX map for ease of interpretation. The same trend is clearly visible on the linear profile of the EDX signal in Figure 3g. Indeed, the atomic ratio Bi:Sb:Te:Se in region 2 significantly differs from the composition \( Bi_{1.3}Sb_{1.7}Te_{0.9}Se_{0.6} \) which obtained for ordered TI layers. At the same time, the composition of the amorphous diffuse layer (region 3) is equal to \( Bi_{1.4}Sb_{0.9}Te_{1.7}Se_{1.2} \) and is very close to the nominal composition observed in the TI. Thus, the bright contrast in region 2 is primarily associated with the enrichment of this region with the heaviest bismuth compared to other elements.

We associate region 2 with \( S' \). This region can be either proximitized by \( S \) or it can have intrinsic superconductivity. Usually, superconducting states just below SC electrodes stem from the superconducting proximity effect. However, TEM results suggest other origins, such as intermixing of the elements, strain, and other composition issues. For example,
Bi$_3$Sb$_4$ alloys are known to be superconducting\cite{35,36} and region 2 is enriched in Bi and Sb. In addition, it is known that tetradymite topological insulators become superconducting under pressure. The superconducting phase transition is accompanied by two structural phase transitions, according to Ref. \[37\]. Therefore, we suggest that $S'$ comes from the fact that $I_{c2}$ has a linear dependence on the external magnetic field (see Figure 2b and Section S2, Supporting information), which is typical for thin films of superconductors.\cite{38} $I_{c2}(T)$ also supports this hypothesis. It was shown that if $\Delta^\prime_{\text{Nb}} \gg \Delta^\prime_{\text{Tl}}$ (i.e., $T^\prime_{\text{cNb}} \gg T^\prime_{\text{cTi}}$), the temperature dependence of the critical current of the $SS'$ interface in the KO-1 model is determined by the temperature dependence of $\Delta^\prime_{\text{Tl}}$, which can be approximated by the square root.\cite{19,39} In our case, $I_{c2}(T)$ is well fitted by the square root dependence (see Figure 2a and the Section S2, Supporting information), which is another argument for intrinsic superconductivity of $S'$. Therefore, in our case, the second step in the $I$–$V$ curve is associated with the destroying of the superconductivity in the $S'$ region.

We believe that the $S'$ region is formed during the Ar-plasma etching, performed before the Nb deposition. It explains why Region 2 is enriched in Bi and Sb, since it is known that Se and Te are more prone to sublimation, compared with Bi and Sb. In addition, plasma treatment can change the crystal structure of the material, as shown, for example, in Ref. \[40\]. In contrast, the direct deposition technique just after cleaving crystals allows to obtain a clean interface without an additional phase, akin to Ref. \[41\]. Nevertheless, an additional study is required to understand the exact impact of the Ar-plasma on the surface of the topological insulator.

4. Conclusion

Thus, we have demonstrated the existence of two critical currents in SQUID based on Fe-doped TI BiSbTe$_2$Se. We analyzed the dependencies of these critical currents on temperature and magnetic field and carried out structural and elemental analyses of the Nb interface with Fe-doped TI BiSbTe$_2$Se. The conducted study shows that the detected effect can be explained by the presence of intrinsic superconductivity in the transition layer between Nb and the Fe-doped TI BiSbTe$_2$Se. Apparently, this layer is formed during the etching of the TI surface and subsequent deposition of Nb. If so, we are raising serious concerns about this common approach for device fabrication. We claim that even in the case of the high $S$–TI interface transparency, determined from the $I_{c}(T)$ dependence, the interface may have complex structure, which can adversely affects the physics of artificially designed topological superconductors, including Majorana-related effects. This complex structure of the interface should be taken into account when the technological process involves Ar-plasma cleaning. If the atomically flat and clean interface is needed, we suggest using deterministic transfer inside the glove box,\cite{42–46} in situ methods,\cite{41,47} or other technological processes.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

V.S.S. conceived the project and supervised the experiments, A.F. and D.V.V. performed the ARPES measurements, E.K. did mechanical exfoliation, A.G.S. and A.K. realized e-beam lithography, V.S.S. and R.A.H. provided the explanation of the observed effects, I.B. and R.A.H. constructed the RSJ-model, I.B. did numerical modeling, S.N.K did the fitting by the KO-2 model, A.K. wrote the manuscript with the contributions from other authors.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

interfaces, superconductivities, transmission electron microscopy, topological insulators

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