Proximity effects at the interface of a superconductor and a topological insulator in NbN-Bi$_2$Se$_3$ thin film bilayers

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Abstract

In a search for a simple proximity system of a topological insulator and a superconductor for studying the role of surface versus bulk effects by gating, we report here on a first step toward this goal, namely the choice of such a system and its characterization. We chose to work with thin film bilayers of grainy 5 nm thick NbN films as the superconductor, overlayed with 20 nm thick topological layer of Bi$_2$Se$_3$ and compare the transport results to those obtained on a 5 nm thick reference NbN film on the same wafer. Bilayers with ex situ and in situ prepared NbN-Bi$_2$Se$_3$ interfaces were studied and two kinds of proximity effects were found. At high temperatures just below the superconducting transition, all bilayers showed a conventional proximity effect where the topological Bi$_2$Se$_3$ suppresses the onset or mid-transition $T_c$ of the superconducting NbN films by about 1 K. At low temperatures, a cross-over of the resistance versus temperature curves of the bilayer and reference NbN film occurs, where the bilayers show enhancement of $I_c(0)$, the supercurrent, and the Andreev conductance, as compared to the bare NbN films. This indicates that superconductivity is induced in the Bi$_2$Se$_3$ layer at the interface region in between the NbN grains. Thus an inverse proximity effect in the topological material is demonstrated.

Keywords: topological superconductivity, proximity effects, topological insulator--superconductor bilayers

(Some figures may appear in colour only in the online journal)

1. Introduction

Topological superconductors (TOS) are interesting due to the expected zero energy Majorana states at vortices [1, 2], and their potential application in quantum computing [3]. Bulk TOS such as copper doped Bi$_2$Se$_3$ should have been the simplest materials to study TOS properties, but complications due to their inherent inhomogeneity [4] and the presence of possible superconducting impurity phases such as CuSe$_2$ [5], make them less attractive for such investigations. Another way for investigating TOS is by inducing superconductivity in a topological insulator or in semiconductor-nanowires with strong spin–orbit coupling via the proximity effect (PE) [6–9]. Unconventional superconductivity in these systems, such as reflected by the observation of zero bias conductance peaks (ZBCP), indicates the presence of zero (or near zero) energy bound states that might be due to Majorana zero energy modes. These Majorana modes are strictly located at zero energy while standard Andreev bound states (ABS which also show ZBCPs) can originate also in very close to zero energy states. Thus reducing the thermal broadening of the ZBCPs by further lowering the temperature should help to distinguish between them, but even then, ABS at strictly zero bias could not be ruled out [10]. Another complication involves the spatial sharpness of the boundary region between the superconductor and the topological or semi-conducting material. It was shown that a smoothly varying boundary leads to near-zero-energy end states even in the topological trivial case [11]. Since this boundary is generally created by gating in the experiments, its smoothness and tapering add more...
uncertainty to the interpretation of the observed ZBCPs as due to Majorana modes [8, 9]. The role of gating in these nano-wire-superconductor experiments was further investigated recently and a variety of additional phenomena such as ZBCP oscillations versus gate voltage and magnetic field were observed and interpreted in the context of Majorana modes as well as alternatives such as Kondo and disorder effects [12]. Hence, gating is sufficiently important in these studies and we decided to look for a simple proximity system of a topological insulator and a 2D superconductor for studying the role of surface versus bulk effects by gating. We extended our previous investigations of Bi$_2$Se$_3$-NbN junctions [13, 14] to bilayers of this system using ultra-thin, grainy NbN layers with weak-links in between the grains, overlayed with a thicker Bi$_2$Se$_3$ layer which facilitates stronger-links between the NbN grains via the (inverse) PE. Here we report on transport characterization of this system, while gating studies of these bilayers will be performed in the future.

2. Preparation and characterization of the films and bilayers

The NbN and Bi$_2$Se$_3$ thin films were prepared by laser ablation deposition using the third harmonic of a Nd:YAG laser. The NbN films were deposited using a metallic Nb target under 30–40 mTorr of N$_2$ gas flow at 600 °C heater block temperature, while the Bi$_2$Se$_3$ layers were deposited using a highly Se-rich target (pressed at room temperature with Bi:Se atomic ratio of 1:17), under vacuum and at 300 °C. The laser was operated at a pulse rate of 3.33 Hz, with high fluence for the deposition of the NbN films (≈10 J cm$^{-2}$) and low fluence for the deposition of the Bi$_2$Se$_3$ layers (≈1 J cm$^{-2}$). All films were deposited on fused silica wafers of 10 × 10 mm$^2$ area. X-ray diffraction measurements of our typical bilayer of 20 nm thick Bi$_2$Se$_3$ on 5 nm thick NbN on a fused silica wafer, showed that the Bi$_2$Se$_3$ cap layer grew with preferential c-axis orientation normal to the wafer with $c = 2.85$ nm. Figure 1 shows atomic force microscopy images of the surface morphology of the as deposited NbN film (a) and the bilayer (b), together with their corresponding typical line profiles (c) and (d), respectively. The rms roughness of the film is ≈0.5 nm and that of the bilayer is of about 2 nm. We note that a 10 nm thick NbN layer on (100) SrTiO$_3$ wafer was much smoother with RMS roughness of only 0.1 nm (not shown). Since grainy thin films are essential for the present study (otherwise a superconducting short will mask all our transport data), we chose to use the fused silica substrates on which the films are much rougher as seen in figure 1. SEM images of similar NbN films on glass showing their grainy nature can also be seen in [15]. We comment here that for the goal of observing Majorana fermions in the present system a

![Figure 1. Atomic force microscope (AFM) images of a 5 nm thick NbN film (a) and of a 20 nm Bi$_2$Se$_3$ on a 5 nm NbN bilayer (b) together with their corresponding line profiles along the lines shown in (a) and (b), as depicted in (c) and (d), respectively.](image-url)
few changes will have to be made. A geometry similar to that proposed by Fu and Kane [2] will have to be used. The bulk conductivity of the Bi$_2$Se$_3$ layer will have to be further reduced, possibly by gating, to avoid the three-dimensional character of the induced superconductivity. Moreover, for detecting Majorana fermions one might need even thinner films, thus less grainy films would have to be used, such as can be grown on SrTiO$_3$ wafers. In the present study however, since we are looking for PE only, we shall use the more grainy and thicker bilayers and films as described earlier.

Two types of bilayers were used in the present study. One using an ‘ex situ’ process where the NbN film was exposed to ambient air for 1 h after which it was measured under He atmosphere, and then immediately put back into the vacuum chamber for the deposition of the Bi$_2$Se$_3$ cap layer. The other was an ‘in situ’ process where both Bi$_2$Se$_3$-NbN bilayer and reference NbN film were prepared in the same deposition run without breaking the vacuum, on two halves of the wafer by the use of a shadow mask, similar to that described in [16]. It is well known that NbN films exposed to ambient air develop within several minutes a surface oxide layer of about 1 nm thickness, while oxidation in between the grains takes longer [17]. It was thus essential to investigate both ex situ and in situ bilayers in order to see how the NbN$_x$O$_y$ oxides affect the results. Transport measurements were done by the use of an array of 36 gold coated spring loaded spherical tips for the four-probe measurements on nine different locations on the wafer. Figure 2(a) depicts a schematic drawing of an in situ prepared sample with the bilayer and reference NbN film (separated by a scratch) and the 36 contact locations. Also shown is a representative current and voltage wiring scheme which is switched from contact C1 to contact C10 using an electronic switching box. In the present study, contacts C1, C3, C5 (on the scratched area) and C9 were disconnected, but by reversing the sample position in the measuring probe, most of the shown contact locations could be measured (in reversed order C(i) is C(11-i)). Figure 2(b) shows a schematic diagram of the bilayer cross-section. Three separated NbN grains are drawn here together with their thin native oxide layer (black), and the thin Bi$_2$Se$_3$ layer near the interface in which proximity induced superconductivity occurs (red).

3. Results and discussion

3.1. Ex situ prepared bilayers

Figure 3 shows the resistance versus temperature results of a 5 nm thick NbN film that was exposed to ambient air for 1 h, together with the results measured on a bilayer created on it by the deposition of additional 20 nm thick Bi$_2$Se$_3$ cap layer. Also shown is the same $R$–$T$ data of the NbN film normalized at 8 K to that of the bilayer. One can see that at high temperatures just below the transition, the Bi$_2$Se$_3$ cap layer suppresses $T_c$ of the NbN film in the bilayer by 0.8 K at mid-transition and by 1.2 K in the onset regime at 0.9R(8 K). This indicates a conventional PE like in normal–superconductor

Figure 3. Resistance versus temperature of a 5 nm NbN film after its exposure to ambient air for about 1 h, and of a bilayer of 20 nm Bi$_2$Se$_3$ deposited on this film. Also shown is an $R$ versus $T$ curve of the 5 nm NbN film normalized to that of the bilayer at 8 K. The inset shows a zoom-in on low temperatures.
junctions, where the normal electrons from N penetrate S while suppressing the order parameter in it and therefore also \( T_c \). It turns out that the topological insulator here behaves like a normal metal. This is not surprising since there is still a significant bulk contribution to the \( BiSe_2 \) conductance (in addition to the surface one), though we reduced it by two orders of magnitude as compared to our previous study [6] by the use of a Se rich target. The resistivity of the bilayer in figure 3 (5 m\( \Omega \) cm at 8 K) is mostly due to the 20 nm thick \( BiSe_2 \) cap layer, which corresponds to an electron density of about \( 10^{17} \, cm^{-3} \) [18]. At low temperatures, a cross-over of the (un-normalized) resistance curves of the reference film and bilayer occurs at 3.6 K, and at 2 K the ratio of resistances is \( \sim 6.2 \). The corresponding ratio of the normal state resistances at 8 K is 4.6. Thus, the bilayer resistance due to the \( BiSe_2 \) cap layer in between the NbN grains is reduced by more than expected from the normal resistance values, and this is a sign of a PE in the \( BiSe_2 \) layer at the interface which will be further investigated in the following.

Figure 4 shows the \( I-V \) curves of the bilayer and NbN film of figure 3. Both reveal small critical currents with serial resistance. The inset to figure 4 shows a typical conductance spectrum of the NbN film at 1.9 K and zero field. It reveals a supercurrent peak with finite resistance at low bias, a broader Andreev structure up to about 8 mV, and supercurrent dips at ±10 and ±25 mV. All these demonstrate the weak-link character of the contacts between the NbN grains in this film. The serial resistance could originate in the thin oxide layer in between the grains or the intrinsic inter-grain resistance in an ultra thin film. The large width of the main peak can be due to supercurrent distribution in the grains, and the broader than the gap Andreev structure to a few weak-links connected in series between the voltage contacts (see figure 2(a)). The supercurrent dips are known to originate in heating effects [19], and here they just show again the presence of supercurrents. Figure 5 shows the conductance spectra of the bilayer of figure 3 at 1.84 K under various magnetic fields. With increasing field the main peak decays slowly, while the Andreev structure decays fast and disappears already at a 0.3 T field. Again, this is a signature of weak-links which are stronger in the bilayer than in the film. This effect will be further enhanced in the \( in situ \) bilayers as we shall see next.

3.2. \textit{In situ} prepared bilayers

Figure 6 shows the resistance versus temperature of an \( in situ \) prepared bilayer of 20 nm \( BiSe_2 \) on 5 nm NbN (shown are the C2 and C4 results) and of the C6, C7 and C8 contacts on the fresh 5 nm NbN film normalized to that of the bilayer at 8 K.

The lower absolute \( T_c \) values in figure 6 as compared to those of figure 3 are due to the higher \( N_2 \) gas pressure used in the...
deposition process (40 versus 30 m Torr, respectively). Nevertheless, at high temperatures the same kind of conventional PE is observed where the $T_c$ values of the bilayer are lower by 0.7–1.2 K as compared to those of the reference NbN film. At low temperatures however, the inverse PE in the Bi$_2$Se$_3$ cap layer is much more pronounced in the in situ deposited bilayer than in the ex situ one. This is demonstrated in the main panel of figure 7, where the bilayer resistance drops to zero at $T_c \sim 2.3$ K while the reference NbN film remains resistive. The noisier data of the NbN film is due to the poor quality of the voltage contacts to this ultrathin and grainy film. The inset to figure 7 shows the corresponding $I$–$V$ curves at 1.82 K of the bilayer and reference film of the main panel. The critical currents of both are much higher here than in figure 4, due to the oxide-free interface in the bilayer, and to the minimal oxide layer in the fresh NbN reference film (after about 10 min exposure to ambient air). Figure 8 shows the conductance spectra of these bilayer (inset) and reference NbN film (main panel). One can see that the supercurrent peaks are much narrower now and the residual resistance is due mostly to flux flow. Moreover, the zero bias conductance of the bilayer is higher by a factor of $\sim 17$ as compared to that of the reference NbN film. These observations together with the Andreev behavior at higher bias indicate that strong-links are established in between the grains of the NbN layer in the in situ bilayers, originating in a strong inverse PE phenomenon. This can be further explained by the scheme of figure 2(b), where three separated NbN grains are shown. In the left hand side contact between two of these grains, the proximity regimes in the Bi$_2$Se$_3$ cap layer overlap thus leading to a strong contact with $I_c > 0$. If this kind of contacts percolate between the corresponding voltage contacts, a strong-link behavior is seen. Similar results were obtained previously in the cuprates [20, 21].

Figure 7. Zoom-in on the low resistance versus temperature regime of the bilayer and reference NbN film of figure 6. The inset shows the corresponding $I$–$V$ curves at 1.82 K and zero field.

Figure 8. Conductance spectra at 1.82 K and zero field of the 5 nm NbN film of figure 7. The inset shows the conductance of the corresponding bilayer of figure 7 together with the data of the main panel for demonstrating its small magnitude.

Figure 9. Resistance versus temperature of fresh in situ prepared Bi$_2$Se$_3$-NbN bilayer and reference NbN film on a different wafer. The data shows very small scattering of the bilayer results with $T_c(R = 0) = 2.2–2.3$ K, while the reference film in this special and rare case shows a very large scattering of the data with $T_c(R = 0)$ of 2.4 K for the C2 contact and no $T_c(R = 0)$ at all for the C4 contact. The inset shows the tunneling-like conductance spectrum of the C4 contact.

We shall now focus on scattering of the data on different wafers. As we have seen in figure 6, scattering of the data was quite small for both bilayer and reference NbN film. This however, was not always the case as figure 9 shows. While the in situ prepared bilayers with their protected interface have $R$ versus $T$ data very similar to that of figure 6, the C2 and C4 contacts on the reference NbN film in figure 9 have very different properties. In this quite rare case, C2 is properly superconducting with $T_c$ of about 2.4 K, while C4 is clearly resistive down to 1.8 K. The inset to figure 9 shows tunneling-like behavior in the conductance spectrum of the C4 contact, indicating its weak-link behavior at low temperature. The different behaviors of these two contacts reflects the fact that there was a good superconduction percolation path between the NbN grain of the film in C2, but not in C4 where discontinuities (or cuts) between the voltage contacts occurred. These apparently originated in defects in the film in the C4
location, which should have been sub-nano meter in size, as they could not be detected using the atomic force microscope. Referring again to the scheme in figure 2(b), one can see that the right hand side contact between the NbN grains has no supercurrent ($I_s = 0$). Thus, if a series of this kind of contacts cut the superconducting percolation path between the NbN grains, the result is a resistive contact, like C4 in figure 9.

Another parameter that affects our results is oxidation or ‘aging’ in ambient air. Figure 10 shows this aging effect on the wafer of figure 9 after 72 h exposure to dry ambient air. These $R$ versus $T$ measurements were taken on a reversely inserted wafer, thus the bilayer has the lower contact numbers now, and C7 here is the same contact as C4 in figure 9. The $R$ versus $T$ results of the bilayer seem unaffected, with no aging effects of its protected interface layer. All the contacts of the reference NbN film however, become resistive now, with the one that was already resistive in figure 9 (C4) having the highest resistance (C7 here). Thus oxidation of the NbN grains in the reference film after a long exposure to ambient air renders the inter grain connections resistive. The bilayer interface though is protected against oxidation and could be used in future gating experiments for longer times.

Finally, we describe test results also of bilayers with different NbN layer thicknesses. Using thinner NbN films with 3–4 nm thickness, yielded insulating-like $R$ versus $T$ dependence with only a change of slope at $T_c$ of the grains, similar to what was observed in [21] in the cuprates. So we decided not to work with these films for the PE study. Figure 11 shows the $R$ versus $T$ results under 1 mA bias current of a bilayer of 20 nm Bi$_2$Se$_3$ on 7 nm NbN and 7 nm thick reference NbN film. Basically, the same effects as observed before with the 5 nm thick NbN layer were found, but now since the 7 nm thick NbN layer has stronger inter grain links, the residual resistances at low temperatures of both bilayer and reference NbN film are even lower. At high temperatures, the mid transition $T_v$ of the NbN reference film is higher by about 0.3 K than that of the bilayer (conventional PE), while at low $T$ the bilayer resistance is more than two orders of magnitude smaller than that of the reference film. This can be understood by the marked difference between the supercurrent of the two as seen in the inset of this figure. We stress that all the previous $R$ versus $T$ results were obtained using 0.01 mA bias only (compared to 1 mA in figure 11). In the present case however, under 0.01 mA bias current, both bilayer and reference film had the same $T_v$($R = 0$) of about 2.5–2.8 K, consistent with the $V–I$ curves in the inset to figure 11. Thus, the inverse PE in the Bi$_2$Se$_3$ cap layer is demonstrated also for these thicker NbN layers. Clearly, if even thicker NbN layers would be used, a superconducting short between the NbN grains would mask any PE effect in the bilayers.

4. Conclusions

A study of proximity induced superconductivity in bilayers made of topological Bi$_2$Se$_3$ thin films capping ultrathin superconducting NbN layers was carried out. The transport results of every bilayer were compared to those of its reference NbN film of the same thickness as in the bilayers and prepared on the same wafer. Conventional PE that suppresses superconductivity in the NbN layer of the bilayer was found at high temperatures, just below the superconducting transition. An inverse PE where superconductivity was induced in the topological material was found at low temperatures. We plan to use the present kind of bilayers in future gating experiments.
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