

# High-temperature superconductivity in a topological insulator

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Producing new effects through the combination of different materials has a long history in science and technology. One of the most intriguing recent ideas is the emergence of Majorana fermions when a topological insulator is placed in proximity with a superconductor. Towards this goal, we produced high-temperature superconductivity in the topological insulator  $\text{Bi}_2\text{Se}_3$  via the proximity to  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ . This was achieved through a new mechanical bonding technique enabling the fabrication of high-quality junctions between novel materials, unobtainable by conventional approaches. The results presented here open new directions for fundamental studies in condensed matter physics and enable a wide range of applications in spintronics and quantum computing.

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Novel phenomena in matter are often obtained by spontaneous symmetry breaking. Therefore, much of modern solid-state physics research has focused on tuning materials to new regimes, such as efforts to study non-Fermi liquid states (1), high-temperature superconductivity (2), and Bose-Einstein condensation (3). Lately, a different paradigm for finding new properties in the solid state has emerged, namely through the sudden change in topological invariants rather than the breaking of symmetries. These topological phases of matter have been proposed (4,5,6) and demonstrated experimentally (7,8) to exist at the surface of some materials with strong spin-orbit coupling. These topological insulators (TI) have topologically-protected surface states predicted to reveal various physical properties including axion electrodynamics (9), as well as practical applications including dissipationless spin currents (10) crucial to spintronics (11).

Perhaps most exciting is the proposal to produce the elusive Majorana fermion by combining both concepts - the broken gauge symmetry of a superconductor with the sudden change in topological invariants at the surface of a TI (12). If obtained, the Majorana fermion would usher in a new era for particle and condensed-matter physics, as well as enable novel quantum technologies such as fault-tolerant topological quantum computing (13,14). The original proposal of proximity-induced superconductivity in an undoped bulk TI put forward by Fu and Kane has not been demonstrated experimentally yet. The proposal has nonetheless reinvigorated the long-studied problem of the proximity effect (15), where superconducting and normal materials are affected by being connected with one another. This effect allows combining different properties of the materials that do not coexist in each individual material, e.g. quasi-relativistic dispersion

in graphene and superconductivity (16). Proximity provides a powerful resource of inducing superconductivity locally in any material, and it has been employed recently for realization of new optoelectronic devices (17). Most of the proximity-based experiments, however, have been performed with low critical temperature ( $T_c$ ) superconductors, whereas inducing superconductivity at higher temperatures with high- $T_c$  materials (18) can enable future realizations of more practical hybrid superconductor based devices.

Here we present the first experimental demonstration of proximity-induced high- $T_c$  superconductivity in a TI. The effect is achieved by a newly-developed method of mechanically-bonded planar junctions, with a focus on  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi-2212) and  $\text{Bi}_2\text{Se}_3$ . We characterize the induced superconducting gap in  $\text{Bi}_2\text{Se}_3$  by current-voltage and differential conductance measurements, studying the spectra of Andreev scattering - a process where an incident electron (hole) creates a Cooper pair in the superconductor with the reflection of a hole (electron) resulting in an excess current (19). The Bi-2212/ $\text{Bi}_2\text{Se}_3$  Andreev spectra exhibit both: the proximity-induced superconducting gap within  $\text{Bi}_2\text{Se}_3$  and the Bi-2212 gap reduced by proximity. The utilization of double-gap Andreev scattering spectra to characterize the proximity effect at superconducting-normal (S-N) interfaces is well established in low- $T_c$  superconductors in contact with normal metals (20) or semiconductors (21). Our results show that the induced superconductivity in  $\text{Bi}_2\text{Se}_3$  persists up to at least 85K – a temperature an order of magnitude higher than any previous observations. Indeed, low-temperature superconductivity has been observed recently in TI with very high levels of Cu doping (22,23,24), and in nanostructures with low- $T_c$  superconductor contacts made using complicated fabrication methods (25,26).

We have developed a simple mechanical bonding technique enabling implementation of practical hybrid TI/high- $T_c$  devices. High-quality bulk  $\text{Bi}_2\text{Se}_3$  and optimally doped Bi-2212 crystals, grown by the floating-zone method were cleaved resulting in atomically flat surfaces over large areas, and were subsequently mechanically-bonded (Fig. 1) in a dry atmosphere. These low-resistance tunnel junctions (27) were then probed via tunneling spectroscopy measurements including current and differential conductance versus voltage (Fig. 1). The carrier density of  $\text{Bi}_2\text{Se}_3$  was determined to be  $1.6 \times 10^{19} \text{ cm}^{-3}$  (27) by far infrared (FIR) spectroscopy (Fig. 1 B). The tunneling spectroscopy experiments were performed using four-point probe measurements in a liquid He flow cryostat at different temperatures ranging from 295K to 4.5K. Our mechanically-bonded tunnel junction method was also verified to perform successfully on Bi-2212/graphite junctions, resulting in typical S-N tunneling spectra (27).

The first experiments were performed on low-resistance Bi-2212/ $\text{Bi}_2\text{Se}_3$  junctions with current along the c-axis. DC voltage versus current (I-V) characteristics (Fig 2A) reveal excess current below the Bi-2212  $T_c \sim 85\text{K}$ , as predicted by the Blonder-Tinkham-Klapwijk (BTK) formalism for very low barrier S-N interfaces (19). The observed excess current is a manifestation of Andreev scattering - the key mechanism for the superconducting proximity effect, (28) suggesting the existence of a proximity-induced superconducting region at the Bi-2212/ $\text{Bi}_2\text{Se}_3$  interface. Just below  $T_c$  the excess current  $I_c$  reaches the maximal value at nearly twice that of the normal-normal (N-N) interface (measured for  $T > T_c$ ) due to the current carried by Cooper pairs when Andreev reflection occurs. At lower temperatures, the excess current is reduced, and the spectrum exhibits

additional features. A different measurement - AC differential conductance below  $T_c$   $[dI/dV]_s$ , divided by the normal state conductance  $[dI/dV]_N$  measured at 105K, with  $\sim 8\text{mV}$  resolution, confirms the DC I-V measurement, demonstrating the zero-bias conductance peak due to Andreev scattering below  $T_c$  (Fig. 2 B). At lower temperatures (around 60K), reduction of higher-bias current was observed, and at the same temperatures additional higher-bias features appeared in the differential conductance measurement. These features result from the gap reduction in Bi-2212 and the induced gap in  $\text{Bi}_2\text{Se}_3$ . However, the detailed spectrum structure of these features could be studied only at lower temperatures.

To demonstrate the spectral features more clearly, we performed both DC and AC measurements at lower temperatures – well below  $T_c$ . DC I-V characteristics of the junction (Fig. 2 C) show excess current at lower voltages up to about 13mV with a correspondingly smaller value  $I_s \sim 2.7\mu\text{A}$ . However, two additional step-like features appear near 27mV and 45mV. These features are confirmed by an AC differential conductance measurement (Fig. 2 D), showing a wide conductance feature centered at zero bias between  $\pm 13\text{meV}$ , and peaks at  $\pm 27\text{mV}$  and  $\pm 45\text{mV}$ , in good quantitative agreement with the DC measurement (Fig. 2 C inset). As discussed below, this conductance spectrum is a clear signature of a proximity-induced superconducting region in the  $\text{Bi}_2\text{Se}_3$ .

When proximity effect occurs (Fig. 3 A (I)), the superconducting gap extends into the normal region ( $\Delta_i$ ) such that near the interface it is reduced from the intrinsic value ( $\Delta_0$ ) to a smaller one ( $\Delta_r$ ). Generally, the superconducting gap is position-dependent along the axis normal to the interface plane,  $\Delta(x)$ , and Andreev scattering probability is

finite in the whole proximity region. However, the scattering can be divided into two main energy ranges: electrons (holes) with energy  $|E| < \Delta_i$  will be mainly Andreev reflected inside the induced superconducting region of the normal material, whereas electrons (holes) with  $\Delta_i < |E| < \Delta_r$  will continue to the S-N interface and Andreev-reflect mainly inside the superconductor near the interface. Thus, conductance spectrum taken on an interface exhibiting the proximity effect will show Andreev scattering features corresponding to both gaps (20, 21). In our experiment, therefore, the central conductance peak is a manifestation of Andreev reflection from the normal region to the proximity-induced superconducting region of  $\text{Bi}_2\text{Se}_3$  with a gap  $\Delta_i \sim 13\text{meV}$ . The reduced gap in Bi-2212 due to the proximity effect appears as a conductance peak at  $\Delta_r \sim 27\text{meV}$ . The change in the zero bias conductance at  $T_c$  is an additional manifestation of the Andreev scattering in  $\text{Bi}_2\text{Se}_3$ . Indeed, just below  $T_c$  the zero-bias conductance increases to nearly twice its value above  $T_c$  (Fig. 3 B). This temperature dependence as well as the spectral shape of the conductance at various temperatures completely rules out any possible heating related effects.

Moreover, a non-uniform junction can have areas with no proximity effect (Fig. 3 A – (II)). These areas will exhibit Andreev scattering with the intrinsic gap of the superconductor, appearing in our experiment as additional peaks at  $\Delta_0 \sim 45\text{mV}$ . The height of the  $\text{Bi}_2\text{Se}_3$  Andreev zero-bias conductance feature is nearly twice the normal conductance value due to the Cooper pair contribution (Fig. 2 D), whereas the width is determined by the induced superconducting gap,  $2\Delta_i$ . The width of the Andreev feature is also manifested in the excess current up to a negative bias  $\Delta_i$  in the DC I-V

characteristic (Fig. 2 C) - nearly twice as high as the current in the normal state (Fig. 2 A). The probability of Andreev scattering inside the Bi-2212 is reduced by the scattering in Bi<sub>2</sub>Se<sub>3</sub>, resulting in slightly smaller contribution to the conductance at  $\Delta_r$ .

For the quantitative theoretical modeling of the effect, the c-axis Bi-2212/Bi<sub>2</sub>Se<sub>3</sub> tunneling spectra were calculated using an extension of the BTK formalism developed by Kashiwaya and Tanaka for anisotropic superconductors (29). In this formalism the quasiparticles in a superconductor are described by a two-component wavefunction

$$\Psi(r) = \begin{bmatrix} f(r) \\ g(r) \end{bmatrix} \text{ corresponding to the electron and hole parts, using Bogoliubov-de}$$

Gennes equations:

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} f(r,t) &= \left[ -\frac{\hbar^2}{2m} \nabla^2 - \mu(r) + V(r) \right] f(r,t) + \Delta(r) g(r,t) \\ -i\hbar \frac{\partial}{\partial t} g(r,t) &= \left[ -\frac{\hbar^2}{2m} \nabla^2 - \mu(r) + V(r) \right] g(r,t) - \Delta(r) f(r,t) \end{aligned} \quad (1)$$

where  $m$  is the electron and hole mass,  $\mu(r)$  is the chemical potential,  $V(r) = H\delta(r)$  is the delta-function barrier potential with the strength determined by  $H$ , and  $\Delta(r)$  is the pairing potential (the superconducting gap). In proximity, the gap is reduced in the superconductor and increased in the normal material (Fig. 3 A (I)), so that the spatial dependence can be described by (21):

$$\Delta(x) = \frac{\Delta_0}{1 + e^{(x+a_N)/\xi_N}} \Theta(x) + \frac{\Delta_0}{1 + e^{(x-a_S)/\xi_S}} \Theta(-x) \quad (2)$$

where  $\Theta(x)$  is a step function,  $\xi_N$  ( $\xi_S$ ) is the normal material (superconductor) coherence length,  $a_N$  determines the induced gap in the normal region, and  $a_S$  determines the gap reduction in the superconductor. In the vicinity of the interface, the

reduced superconducting gap can be approximated by  $\Delta_r \approx \Delta_0 / (1 + e^{-a_s/\xi_s})$  and the induced gap in the normal region is  $\Delta_i \approx \Delta_0 / (1 + e^{a_N/\xi_N})$ . Andreev scattering in the proximity region can be modeled therefore by two successive scatterings from two interfaces: scattering within  $\text{Bi}_2\text{Se}_3$  with the induced gap  $\Delta_i$  followed by scattering within Bi-2122 with the reduced gap  $\Delta_r$ . The calculated spectra with the modified gaps as fit parameters show good agreement with the experimental conductance measurements (Fig. 3 D).

To further verify that the Andreev scattering is due to the proximity induced superconductivity in  $\text{Bi}_2\text{Se}_3$ , and dismiss any possible processes occurring in the Bi-2212 alone, the Bi-2212/ $\text{Bi}_2\text{Se}_3$  junction barrier was increased mechanically. As expected, differential conductance measurement performed on the high-resistance junction revealed only spectra typical of a c-axis tunneling measurement on Bi-2212 (30), indicating the lack of a proximity effect. Specifically, the superconducting gap of Bi-2212 was observed, however no Andreev peak appeared at the Bi-2212 reduced gap  $\Delta_r$ , and no conductance features corresponding to proximity-induced gap in the  $\text{Bi}_2\text{Se}_3$  (Fig. 3 C). The differential conductance spectrum for the high-resistance junction showed the superconducting gap of Bi-2212  $\Delta_0$ , and its temperature dependence agrees with the values from the low-resistance junction measurement (Fig. 3 B inset), confirming the origin of the conductance peaks.

Our results present clear evidence of proximity-induced high- $T_c$  superconductivity in the TI  $\text{Bi}_2\text{Se}_3$ . We achieved this via our newly-developed technique of mechanically-bonded junctions, and confirmed it by various experiments. Specifically,



below  $T_c$  we observe Andreev features corresponding to a reduced gap of Bi-2212 appearing concurrently with an induced gap in  $\text{Bi}_2\text{Se}_3$ . The disappearance of these features in the same junctions when the barrier is increased confirms that proximity effect is the origin of these features. The proximity-induced  $\text{Bi}_2\text{Se}_3$  superconductivity is demonstrated at temperatures at least an order of magnitude higher than previously reported results. The developed mechanical bonding technique may render various future experiments on novel materials, including high- $T_c$ , and TI feasible, and our TI proximity demonstration paves the way for practical realization of TI-based devices involving superconductivity, including Majorana fermion based topological quantum computing.

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### **Figure captions:**

**Figure 1.** (A) Atomic force microscope image of the crystal surface. (B) FIR spectrum of the  $\text{Bi}_2\text{Se}_3$  crystal showing the plasma resonance corresponding to carrier density of  $1.6 \times 10^{19} \text{ cm}^{-3}$ . (C-F) Junction fabrication technique : (C) Bi-2212 crystal is cleaved using scotch tape. (D)  $\text{Bi}_2\text{Se}_3$  is sandwiched between glass slides with double-sided tapes and the top glass slide is lifted off, cleaving a flat surface. (E)  $\text{Bi}_2\text{Se}_3$  transferred to a Cu sample holder, and the cleaved Bi-2212 crystal is applied to  $\text{Bi}_2\text{Se}_3$  using GE varnish. (F) Contacts are made with Ag epoxy. (G) Experimental setup: four-point DC current-voltage and AC differential conductance measurements performed down to 4.5K, using a liquid He flow cryostat with lock-in amplifiers. DC bias from the power supply is combined with the AC signal from the voltage lock-in amplifier in a transformer-based adder.

**Figure 2.** (A) DC current-voltage characteristics of the low-resistance junction for different temperatures above and below  $T_c$ . Below  $T_c$ , excess current typical of Andreev reflection at an S-N interface is exhibited. 70K I-V curve shows the maximal excess current  $I_e \sim 4.85 \mu\text{A}$  – similar to the value of the normal junction (above  $T_c$ ) at the voltage where the superconducting-state current becomes linear (black line). The total excess current is smaller for lower temperatures with I-V data showing additional features. (B) AC Differential conductance  $[dI/dV]_s$ , normalized by the normal state conductance  $[dI/dV]_N$  at 105K, for a low-resistance junction at different temperatures above and

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below  $T_c$ . A zero-bias conductance Andreev peak is clearly seen below  $T_c$ , with additional features in the spectrum appearing at lower temperatures. The curves are shifted for clarity. **(C)** DC current-voltage characteristics of the low-resistance junction for different temperatures well below  $T_c$ . In addition to the excess current, the low-temperature curves exhibit two distinct plateaus corresponding to the dips in the differential conductance measured for AC bias. The black arrow shows the induced  $\text{Bi}_2\text{Se}_3$  gap ( $\Delta_i$ ) with a current twice of that of the normal state at the same voltage (Fig. 2 A), and the red arrow indicates the reduced Bi-2212 gap ( $\Delta_r$ ), while the intrinsic Bi-2212 gap ( $\Delta_0$ ) is shown by purple arrows. Thus, the total excess current  $I_s \sim 2.7 \mu\text{A}$  is smaller for lower temperatures. The inset shows the DC and the AC differential conductance at 4.5K, indicating the good correspondence between the different measurements of  $\Delta_0$  (purple arrow),  $\Delta_r$ , and  $\Delta_i$  (red arrow). **(D)** AC Differential conductance  $[dI/dV]_s$ , normalized by the normal state conductance  $[dI/dV]_N$  at 105K, for a low-resistance junction for different temperatures well below  $T_c$ . The curves are shifted for clarity. The zero-bias conductance feature is due to the Andreev reflection between the normal and proximity induced superconducting regions in  $\text{Bi}_2\text{Se}_3$ , where the width of the peak is nearly  $2\Delta_i$ . The two additional peaks indicate the reduced and the intrinsic Bi-2212 gaps.

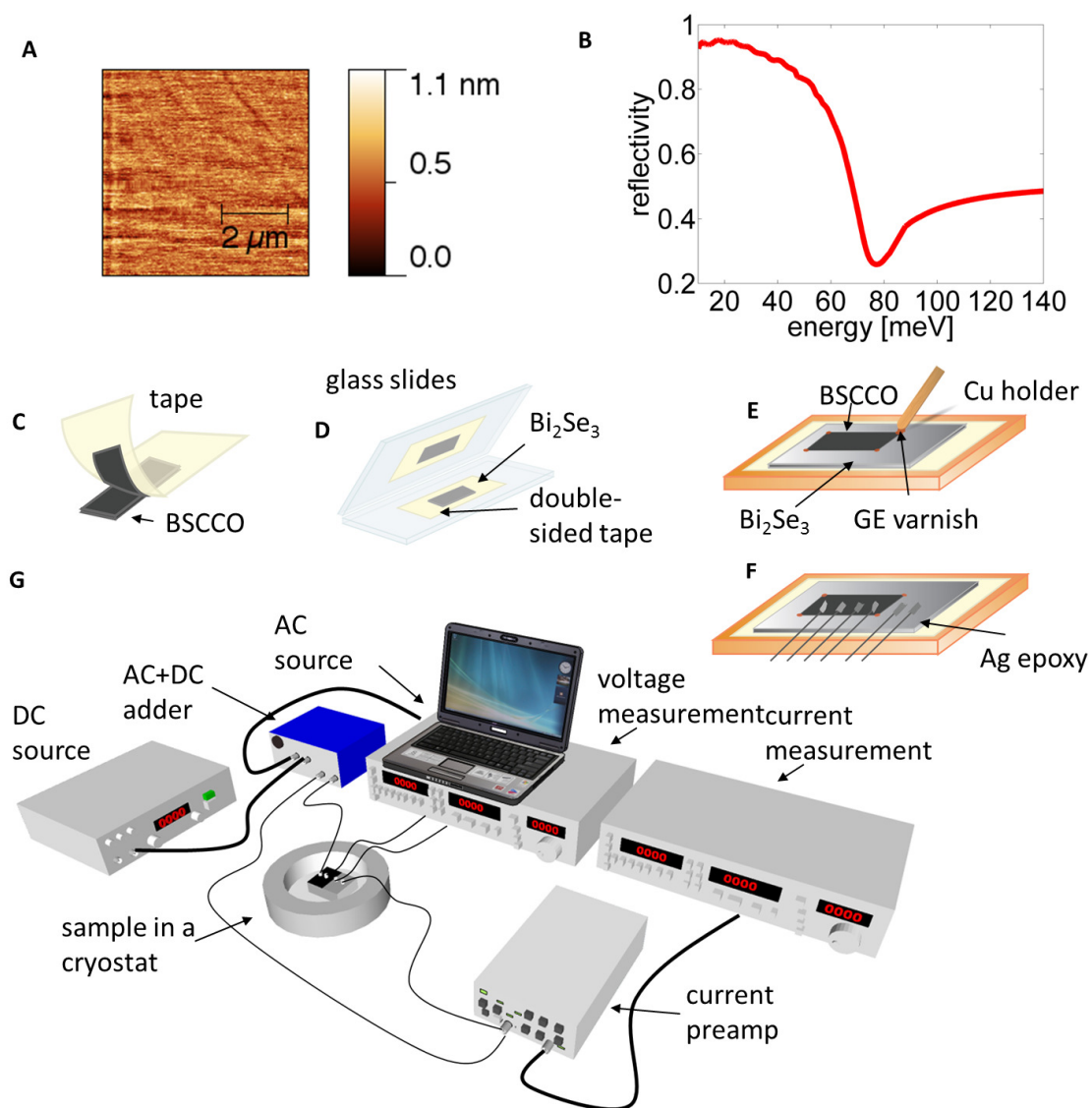
**Figure 3.** **(A)** A schematic drawing of the device in two regimes: **(I)** low-resistance with proximity induced superconductivity in  $\text{Bi}_2\text{Se}_3$ . Andreev scattering takes place in the whole proximity region with lower energy particles reflected mainly in  $\text{Bi}_2\text{Se}_3$ , and higher

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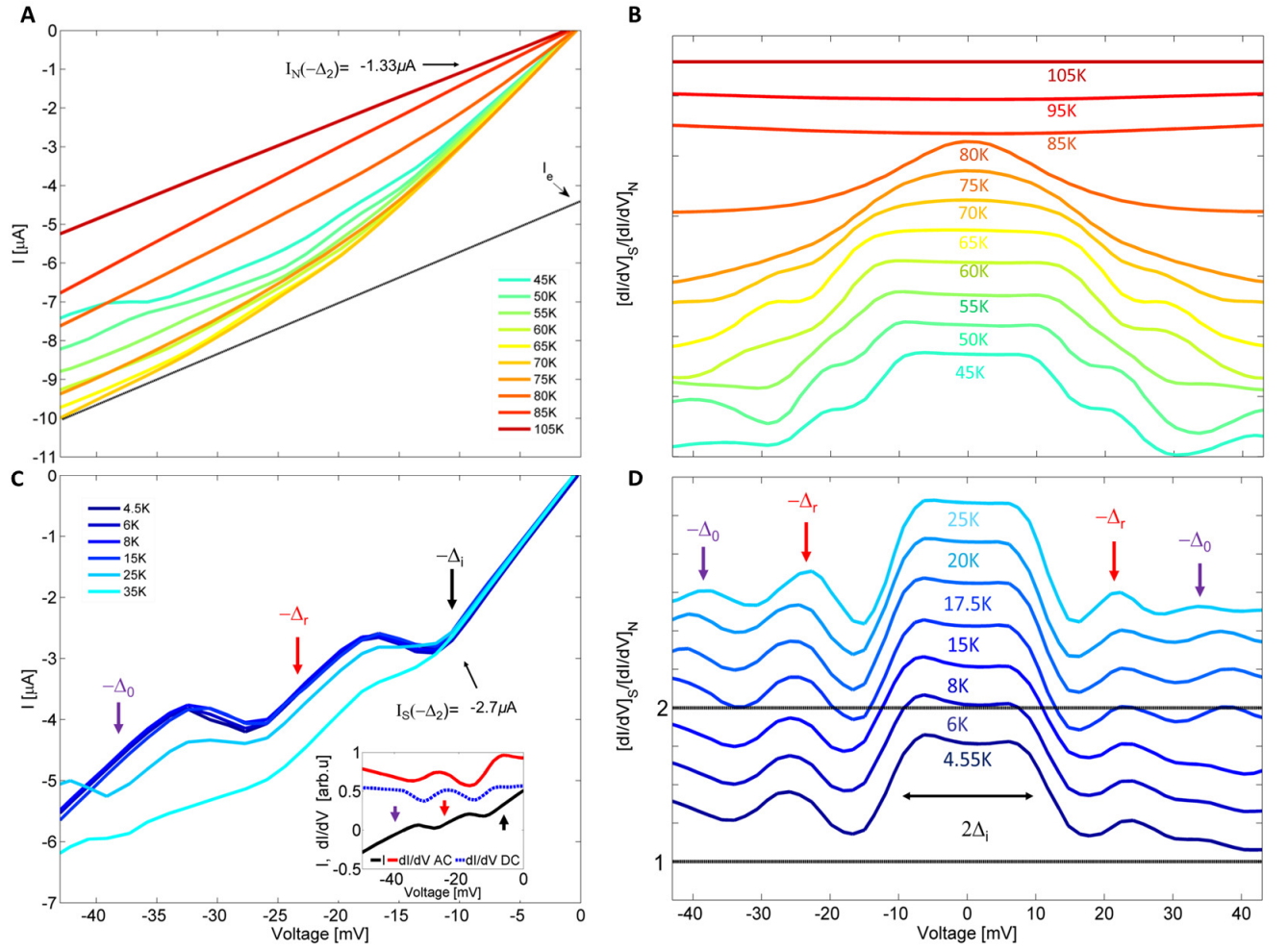
energy ones mainly in Bi-2212. (II) High-resistance with no proximity induced superconductivity in Bi<sub>2</sub>Se<sub>3</sub>. Andreev scattering occurs at the interface. **(B)** Temperature dependence of the zero-bias differential conductance. Below T<sub>c</sub> the Andreev process enhances the conductance by almost a factor of 2. The inset shows the temperature dependence of the reduced Bi-2212 gap  $\Delta_i$  (blue circles) and the intrinsic Bi-2212 gap  $\Delta_0$  in the low-resistance junction (green squares), and in the high-resistance junction (red diamonds). **(C)** Differential conductance  $[dI/dV]_s$ , normalized by the normal state conductance  $[dI/dV]_N$  at 105K, for a high-resistance junction for different temperatures, demonstrating the typical quasiparticle tunneling differential conductance resulting in a conductance dip in the gapped region. The curves are shifted for clarity. The red arrows indicate the intrinsic superconducting gap of Bi-2212  $\pm\Delta_0$  around 45mV.



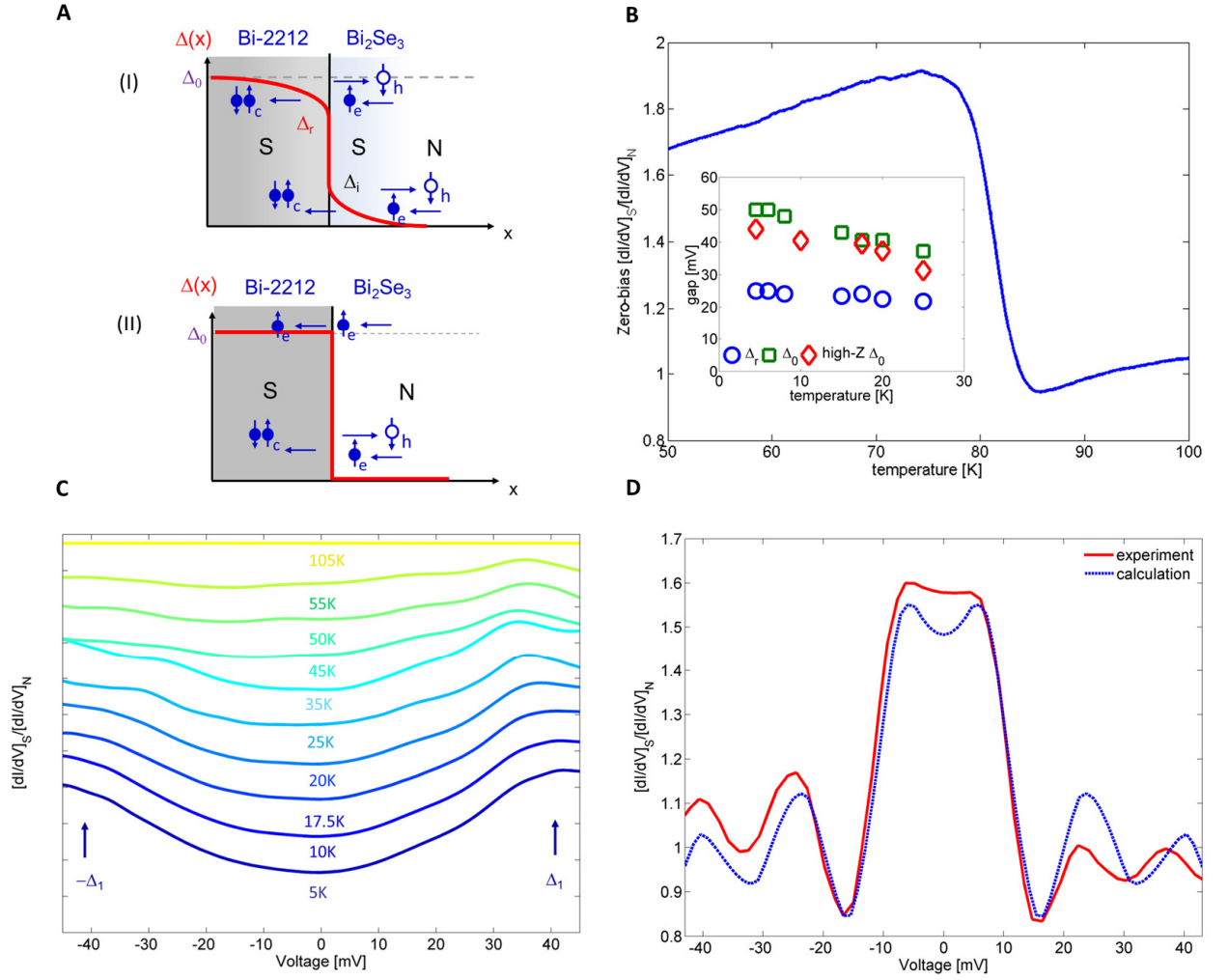
**Figure 1**



**Figure 2**



**Figure 3**



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Supporting Online Material for

**High-temperature superconductivity**

**in a topological insulator**

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**Device Fabrication:** High-quality bulk  $\text{Bi}_2\text{Se}_3$  crystals were prepared as described elsewhere (S1). Additionally, optimally doped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi-2212) crystals ( $T_C \approx 85\text{K}$ ) were grown by the floating-zone method (S2). The carrier density of  $\text{Bi}_2\text{Se}_3$  was determined by far infrared (FIR) spectroscopy. Specifically, reflectance of the (111) surface of the  $\text{Bi}_2\text{Se}_3$  crystal was measured using a modified Bruker Vertex 80V FTIR spectrometer. The sample was mounted on a cone and gold coated in-situ to correct for spurious effects and produce absolute reflectance spectra (Fig. 1 B) (S3). Comparison of the plasma edge at 78 meV with previous  $\text{Bi}_2\text{Se}_3$  FIR spectroscopy data (S4) reveals a carrier density of  $1.6 \cdot 10^{19} \text{ cm}^{-3}$ . Bi-2212/ $\text{Bi}_2\text{Se}_3$  tunnel junctions were fabricated by using

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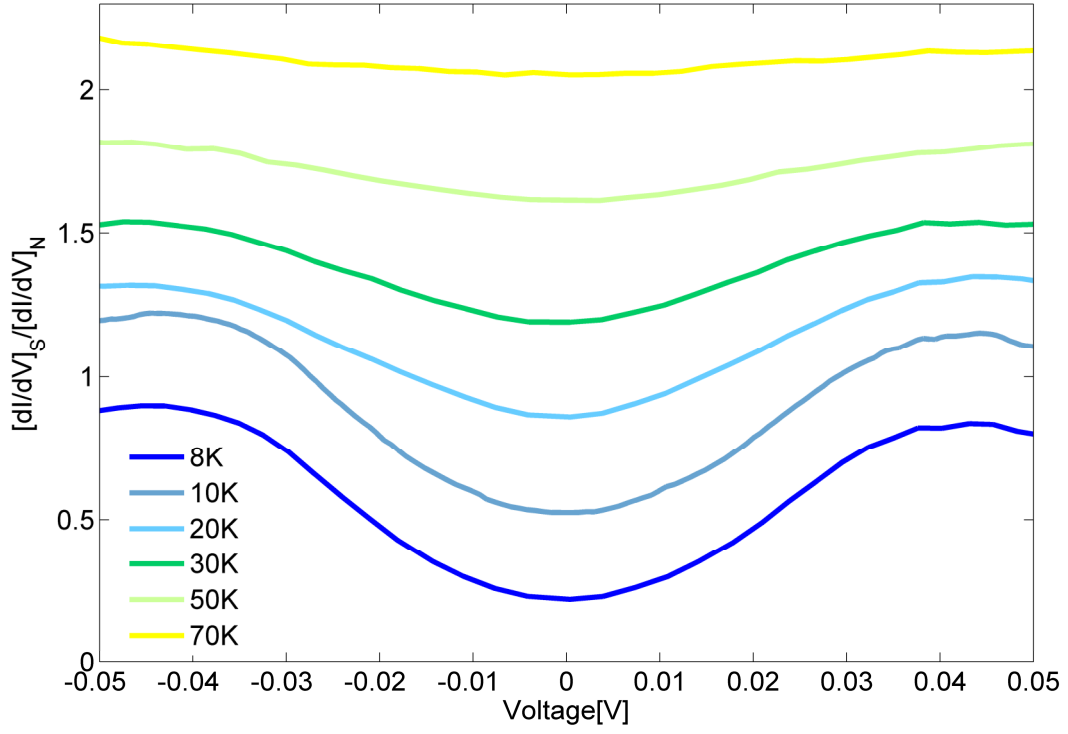
these crystals as the starting materials. Bi-2212 was cleaved in a dry box with adhesive tape (Fig 1. C), and a clean flat piece of Bi-2212 with the thickness of a few hundred  $\mu\text{m}$  was produced. Simultaneously  $\text{Bi}_2\text{Se}_3$  was cleaved in the dry box by being sandwiched between two glass slides with double-sided tapes on both of them. After applying slight pressure to the glass slides, the top glass slide was lifted off (Fig 1. D). This approach was taken due to the difficulty of producing flat surfaces by cleaving  $\text{Bi}_2\text{Se}_3$  with scotch tape, as was done for Bi-2212. Next, the  $\text{Bi}_2\text{Se}_3$  was transferred to a Cu sample holder by attaching a double-sided tape to the Cu and placing one of the glass slides with  $\text{Bi}_2\text{Se}_3$  on it on the sample holder. By lifting off the glass slide, a smooth and fresh surface of  $\text{Bi}_2\text{Se}_3$  was left on the sample holder. The Bi-2212 was then attached to the  $\text{Bi}_2\text{Se}_3$  by placing the Bi-2212 on the  $\text{Bi}_2\text{Se}_3$ , applying GE varnish on the corners of Bi-2212 (Fig 1. E). Contacts were made on the sample using Cu wires and Ag epoxy (Fig 1. F).

**AC and DC Measurements Setup Description:** Four-point probe measurements were performed in a Liquid Helium flow cryostat at different temperatures ranging from 295K to 4.5K. Bias-dependent differential conductance as well as current vs. voltage were measured on the Bi-2212/ $\text{Bi}_2\text{Se}_3$  junction using two lock-in amplifiers (Stanford Research Systems SR810) with a resolution of 8mV, a DC voltage source (BK Precision 1787B), two DC multimeters (Hewlett Packard 3457A & Agilent 34401A) and a home-built, shielded AC+DC adder box (Fig 1.G). One of the lock-in amplifiers was used to produce a small AC voltage output at a frequency of 1.49 KHz. The AC voltage was added to the DC output of a DC power supply using a transformer-based adder. A voltage was applied

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to the sample, with the resulting voltage measured with a lock-in (the AC part) and a multimeter (the DC part). At the same time, the current was converted to voltage and amplified with a current preamplifier (SRS 570) and measured with another lock-in (AC) and a multimeter (DC). Therefore, the AC current of the sample divided by the AC voltage of the sample, which is proportional to the  $dI/dV$  (differential conductance), was measured at different bias voltages.

A Bi-2212/Graphite mechanically-bonded junction was fabricated by the method described above, and AC differential conductance measurements were performed similar to the measurements done on Bi-2212/Bi<sub>2</sub>Se<sub>3</sub>. The graphite-based junction tunneling spectrum measurements show the typical high-resistance superconducting gap (Fig. S1) with the value of  $\pm\Delta_1$  around 45mV – similar to those measured for Bi-2212/Bi<sub>2</sub>Se<sub>3</sub>.



**Figure S1.** AC Differential conductance  $[dI/dV]_s$  normalized by the normal state conductance  $[dI/dV]_N$  at 105K for a Bi-2212/graphite junction at different temperatures below  $T_C$ . The curves are shifted for clarity.

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## SOM References:

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