Supplementary Information for Unconventional Josephson Effect in Hybrid Superconductor-Topological Insulator Devices

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Materials and methods

Single crystals, grown by the binary melt method, of (Bi1−xSbx)2Se3 were synthesized by slow cooling a binary melt of Bi (99.9999%), Sb (99.99%) and Se (99.9999%) starting materials, mixed in the ratio 0.33:0.05:0.62. The actual amount x of Sb in the single crystals is approximately 0.01, as measured in microprobe analysis. Bulk single crystals with dimensions 1×1×0.1mm3 showed a non-metallic resistivity at low temperatures with a bulk carrier density of 7×1016 cm−3 as deduced by Hall and Shubnikov-de Haas analysis.

Bi2Se3 nanowires, synthesized via the vapor-liquid-solid (VLS) method, were grown in a 12-inch horizontal tube furnace with a quartz tube. Bi2Se3 source powder (99.999%, from Alfa Aesar) was placed in the center of the furnace (540°C). The growth substrate, a Si wafer with a thermally-evaporated, 10 nm Au film, was placed at a downstream, lower temperature zone (350°C). High purity Ar gas delivers vapor from the source materials to the growth substrate at 130 sccm for 2 hours with 1 Torr pressure.

Samples of 50-100 nm thickness were prepared by mechanical exfoliation of the above two starting material, using a method similar to exfoliation of graphene [S1]. Prior to sample exfoliation, SiO2(300 nm)/Si wafers were cleaned in acetone and isopropanol. HMDS was deposited on the SiO2 surface in efforts to reduce the amount of H2O contamination. Samples were exfoliated using Nitto-Denko tape and immediately after exfoliation PMMA 950 A4 (Microchem Crop.) was spun on the chip for 60 secs at 4000 rpm, followed by a baked for 30 mins at 180 °C on a hotplate. After electron beam exposure of the resist and development in MIBK/IPA 1:3, a 3 min UV-ozone was used to remove residual PMMA in the exposure contact areas. Ion milling of the exposed contact areas was performed to obtain low contact resistance to the Bi2Se3 flakes. The recipe of the ion mill is a 300 V beam voltage, 30 mA beam current producing an ion current density of 0.1 mA/cm2, 60 V acceleration, 12 sccm argon flow, and a 10 secs exposure. Immediately following ion milling, the samples were loaded into an e-beam evaporator equipped with oil-free pumps, and Ti/Al (3 nm and either 60 nm or 100 nm) were evaporated at a rate of 1 Å/s onto the sample at a pressure of ~5×10−8 Torr at the start of each evaporation.

Samples were measured in a 3He/4He dilution refrigerator with a base temperature of 12 mK. To reduce the effects of thermal radiation, three stages of filtering were employed: room-temperature LC π-filters, a 3-pole RC filter anchored to the mixing chamber designed to filter in the frequency range of 1 MHz to 5 GHz [S2], and 10 kΩ “on-chip” resistors used in conjunction with the inline capacitance of the twisted-pair wiring to produce a low-pass, RC-filter of cutoff frequency ~50 KHz. All experiments were performed in an electrically- and acoustically-shielded room. Device DC resistance and differential resistance were measured using a standard DC and lock-in technique. The applied AC current was kept low enough such that the AC voltage across the device never exceeded 500 nV, below the base temperature of the fridge k_BT~1 µeV. To obtain accurate magnetic field values, a low-noise Yokogawa 7651 was used to supply the current to the superconducting magnet, producing a <0.3 µT noise level in the magnet.

The transport properties of the exfoliated binary melt samples were characterized by making a device with contacts in a Hall-bar geometry and Bi2Se3 thickness of 100 nm. The extracted density, mobility and diffusion constant D of these samples are 7.8×1017 cm−3, 1950 cm2/Vs, 0.02 m2/sec, respectively. We note that these values reflect merged contributions from both the surface and bulk, where it is expected that the surface electrons have a much higher mobility [S3]. Hence, the values for mobility and D are a lower bound for the values of the surface electrons.

To extract the superconducting and normal state properties of the leads, four samples were prepared with the same recipe as the leads of the JJ devices: Ti/Al of thickness 3 nm/(60, 100 nm), both of (L, W)=(10, 1) μm. Each thickness of Al was placed either directly on the SiO2 substrate or on top of a Bi2Se3 flake. Characterization of these test samples was performed via a four-terminal resistance measurement at room temperature.
(290 K), 4 K and base temperature of the dilution refrigerator (12 mK). Little variation was found in the superconducting properties of the Al patterned on the SiO$_2$ substrate and on Bi$_2$Se$_3$. The resistivity of the samples at room temperature and 4K were $3.0 \times 10^{-8}$ Ω m and $0.3 \times 10^{-8}$ Ω m, respectively. The critical currents and magnetic fields for the 100 nm (60 nm)-thick Al-films were 488 μA (208 μA) and 11 (13) mT respectively.

Characteristic lengths and regimes for JJs

There are many different regimes in JJs that produce different behaviors. The regime applicable to the device is determined by the length of the junction $L$ relative to five intrinsic length scales: coherence length of the Al leads $\xi_A=1.6$ μm; the superconducting coherence length of the metallic (Bi$_2$Se$_3$) weak link $\xi_N$, and three properties of the (Bi$_2$Se$_3$) weak link unrelated to the superconductor: the thermal length $\ell_T$, the mean-free path $\ell_e$ and the phase-coherence length $\ell_\varphi$. Several of these lengths depend on the diffusion constant. We use the merged value for $D$ as noted above (see below for more comments on this). $\xi_N$ can be calculated from the diffusion constant by $\sqrt{\hbar D/\Delta}$, where $\Delta$ is the superconducting gap of Al, which we calculate from the BCS equation $\Delta=1.76 k_B T_C$, which for a $T_C$ of 1 K (a typical value for our leads) is 151 μeV. This produces a value for $\xi_N$ of $\sim 280$ nm. $\ell_T=\sqrt{\hbar D/2\pi k_B T}$, which at 12 mK is $1.3$ μm. $\ell_e$ can be extract from D as $D=1/2 \nu_F \ell_e$, giving $\ell_e=80$ nm, where we have used the value of $\nu_F=4.2 \times 10^5$ m/s from ARPES measurements [S4]. $\ell_\varphi$ was estimated from the half-width at half-max of the weak antilocalization correction to the longitudinal resistance (measured in the Hall bar) to be 650 nm. We note that $\xi_N$, $\ell_T$, $\ell_e$ and perhaps $\ell_\varphi$ all represent lower bounds since we have used the average value of $D$ from the surface and the bulk. The $D$ for the surface electrons is expected to be higher.

For the device should be compared to the Josephson penetration depth $\lambda_J = \sqrt{\Phi_0/(2\pi \mu_o J_C (2\lambda_L + L))}$, where $\lambda_L$ is the London penetration depth $\sim 50$ nm [S5]. We place a lower bound on $\lambda_J$ by using the junction parameters that would make it the smallest, i.e. using the largest measured value of $j_C$ and the longest value of $L$: producing a value for $\lambda_{J_{min}}$. This puts our junctions in the regime $1/\ell_e < \xi_N, \xi_A, \ell_\varphi, L_T$ and $W < \lambda_J$, i.e. the short junction, quasi-ballistic regime. In this regime, $I_{C} R_N$ is expected to be described by either the KO-1 or KO-2 theory [S6] and should be independent of $W$: neither of these expectations matches our data.

Additional features in the Differential Response $dV/dI$ as a function of T and B

The differential response $dV/dI$ for a $(L, W)=(50$ nm, 0.9 μm) device as a function of fridge temperature $T_f$ suggests that both the surface and entire bulk should experience the proximity effect. We expect, however, that the supercurrent carried by the bulk to be smaller than that of the surface for two reasons. First, the Cooper pairs have to travel a longer distance from left to right lead through the bulk than through the surface. We can estimate this additional length by assuming the Cooper pairs have to go on average a distance $t/2$ from the left lead to the bulk, then another $t/2$ going from the bulk to the right lead. This makes the effective length of the device for the bulk states to be $L_{bulk}=L+t$. The second is the lower mobility of the bulk, producing a smaller value of $D$. For the device in Fig. 3 of the main manuscript $L=55$ nm and $t=95$ nm, making $L_{bulk}=150$ nm. To calculate the reduction of the critical current, we assume $D$ for the bulk states is 10 times smaller than the calculated value (a factor 12 has been experimentally determined [S7] for the reduction of $D$ for the bulk), giving a $\xi_{bulk}^{bulk} \sim 65$ nm. This estimate yields a bulk critical current of order five times smaller than that of the surface state [S6].

Mechanisms for the reduction of $I_{C} R_N$

We next consider noise as a mechanism for the reduction of $I_{C} R_N$. A typical reduction comes from thermal noise, where the effect on $I_{C} R_N$ depends on the type of junction (overdamped or underdamped). The type of junction is determined the quality factor $Q = \sqrt{2 \pi C_{J} / \hbar \xi_{N}}$, where $C_{J}$ is the capacitance of the JJ. We estimate $C_{J}$ to be 0.5pF using a parallel plate capacitor model between the entire area of the leads of the device and the degenerately-doped Si, yielding $Q=1.2$. This is neither overdamped or underdamped, but at an intermediate value. Numerical evaluation of the $I-V$ characteristics in this regime shows results similar to those of an overdamped junction [S8]. The effect of thermal radiation on overdamped junctions has been calculated [S8], and shown to cause a smearing of the transition between the superconducting and normal state. Since the transitions we measure are still very sharp, we can rule out thermal fluctuations being a source of the reduction of $I_{C} R_N$. We can also calculate what the effect of thermal fluctuations would be had the junctions been in the underdamped regime [S5]: the effective thermal radiation temperature would have to be 3.4 K to reduce $I_{C} R_N$ by the factors we observe. This high temperature is unphysical in our well-filtered setup.

Supercurrent carried by the bulk

The typical thickness $t$ for our devices is 75 nm. The calculated $\xi_N (\sim 280$ nm) is larger than this value, which
is shown in Fig. S1(a), where the device exhibits a sharp transition to $dV/dI=0$ below $T_f=850\,\text{mK}$ of the leads. Above $T_C$, the devices show a metallic temperature dependence of resistance. The differential response $dV/dI(I)$ of this junction, typical for all devices measured, at $T_f=12\,\text{mK}$ is shown in Fig. S1(b). Even aside from the transition from a superconducting state to a normal state, which produce large peaks at $I=I_C$, several features are evident and are indicated by labels 1-4. The first occurs in all devices as either a peak or a sharp rise in $dV/dI$ [like the one shown in Fig. S1(b)], at $V=200\,\mu\text{V}$. The second, broad peak appears at $V=300\,\mu\text{V}$, near $2\Delta/e$ of Al, where a peak in resistance is expected due to the quasiparticle contribution to the resistance from the leads [S9]. The third and fourth peaks are both features occurring above $2\Delta$ of the leads, occurring at $V=500\,\mu\text{V}$ and $V=655\,\mu\text{V}$, respectively. Peaks above $2\Delta$ have also been seen previously in TI JJs [S10].

The effect of $T_f$ on $dV/dI$ is shown in Fig. S1(c). A typical trend is observed, where increasing $T_f$ reduces $I_C$. Additional “shoulder” dips appearing in $dV/dI$ are evident in the $T_f=300\,\text{mK}$ and $400\,\text{mK}$ traces. On the $400\,\text{mK}$ trace, two dips are seen, at $I=500\,\text{nA}$ and $620\,\text{nA}$ respectively.

Additional shoulder features are also seen in $dV/dI$ above $I_C$ when a magnetic field $B$ is applied [Fig. S1(d)]. These are revealed in cuts of constant $B$ in $dV/dI(B,I)$ for the device shown in Fig. 3 of the main text. At $B = 0$, there are two peaks in $dV/dI$ at $I = \pm I_C$, and no other apparent features. At $B=0.75\,\text{mT}$, dips are evident at $I = \pm 1.6\,\mu\text{A}$, $V=12.5\,\mu\text{V}$ above the resistive transition. The single dip occurs for $0.6\leq B\leq1\,\text{mT}$. Two dips appear beginning at $B=1\,\text{mT}$ and persist throughout the entire second and third lobes, although to a much weaker degree in the third lobe. The double-dip is evident in the cut at $B=2.5\,\text{mT}$, occurring at $I=0.9$ and $1.2\,\mu\text{A}$ and a value of $V=7.4$ and $12.1\,\mu\text{V}$. The dips at $B=8\,\text{mT}$ have all but disappeared, remaining weakly at $I=0.5$ and $0.9\,\mu\text{A}$ and $V=6.3$ and $12.1\,\mu\text{V}$. Currently, we have no explanation for these features, which appear in all devices measured.
Andreev Bound States and the Current-Phase Relation

In this section we consider qualitatively the Andreev bound states (ABS) and the resulting current-phase relation (CPR). States in a JJ within the superconducting gap are associated with the coupled electron-hole pairs that are responsible for transferring Cooper pairs from one lead to the other [S11]. These are known as ABS. To describe the CPR in our junction, we consider several different plausible energy spectra for ABS. The energy levels of conventional ABS are $E(\varphi) \propto \pm \Delta \sqrt{1 - T \sin^2(\varphi/2)}$ [S11], where $T$ is the transmission probability of Cooper pairs from left to right lead. From the energy spectrum of the ABS, the current phase relation (CPR) can be calculated as $I(\varphi) \propto \partial E/\partial \varphi$ [S10]. The energy spectrum for the ABS in a conventional JJ with $T=0.9$ is shown in Fig. S2(a) for the occupied (solid) and unoccupied (dashed) ABS states. The corresponding CPR, calculated by differentiating the $E(\varphi)$ plot, is shown in Fig. S2(b), where a saw-tooth behavior is seen, occurring with a period of $2\pi$. Note that for low values of $T$, this CPR becomes the conventional $\sin(\varphi)$ [S11]. For $p + ip$ and TI JJs, the energy of the ABS becomes $E(\varphi) \propto \pm \Delta \cos(\varphi/2)$, producing a zero-energy crossing at $\varphi = \pi$ [S12]. The $p + ip$ ABS spectra matches the conventional energy spectrum of ABS for $T = 1$. The corresponding CPR –assuming a constant ABS occupancy rather than always being in the ground state, i.e. conserved fermionic parity – is $I(\varphi) \propto \sin(\varphi/2)$, resulting in an anomalous $4\pi$-periodic CPR, predicted for $p + ip$ JJs [S12] and TI [S13] JJs. The energy spectrum of the ABS, and the CPR, in this case are shown in Fig. S2(c,d).

In our phenomenological model, the transmission coefficient $T$ depends on $\varphi$, where the 1D charge modes at $\varphi \neq \pi$ have a low value for $T$ – a result of the interactions in 1D [S14] – and the neutral modes have a much higher value for $T$. This $T(\varphi)$ produces a CPR that is peaked at values of $\varphi$ where the zero-energy modes occur. Fig. S2(e) shows an ABS spectrum for a $T(\varphi)$ [inset of Fig. S2(e)], corresponding to $T(\varphi)=1$ for $\varphi = \pi$ and $T(\varphi)=1/10$ for $\varphi \neq \pi$. The exact value for $T$ away from $\varphi = \pi$ can be changed without much change in the results. To connect the high- and low-$T$ regions, we assume that the width of the peak in $T(\varphi)$ near $\pi$ is set by the energy scale $E_C$, the energy associated with momentum quantization in the phenomenological model. This scenario is shown in Fig. S2(g), where the linearized $E(\varphi)$ is calculated from Eq. 4 of Ref. [S15]. Using a value of $E_C= e I_C R_N=31 \mu eV$ for the device in Fig. 3, values of 2.91 rads ($167^\circ$) and 3.37 rads ($193^\circ$) for $\varphi_1$ and $\varphi_2$ are obtained. $T$ then is modeled as a gaussian with a half max at the values $\varphi_1$ and $\varphi_2$. The calculated ABS spectrum for this transmission coefficient is shown in Fig. S2(e), where a transition through $E = 0$ occurs on the scale of $\varphi_1$ and $\varphi_2$ around $\varphi = \pi$. The corresponding $I(\varphi)$ is shown in Fig. S2(f), where the $\varphi$-dependent $T$ produces an anomalous CPR that is peaked around the values of $\varphi$ where the zero-energy modes occur. This CPR is also $4\pi$ periodic. A similar CPR was obtained in Ref. [S16], where a peak in the CPR was also found to be correlated with the zero-energy crossing in the ABS. In Fig. S2(c) and S2(e), it was assumed that fermionic parity was conserved, resulting in a protected crossing of the two states (solid and dashed lines) at $\varphi = \pi$ [S17]. If the parity of the junction is not preserved, an anti-crossing of these two states occurs [inset of Fig. S2(h)], producing a $2\pi$-periodic CPR shown in Fig. S2(h). This $2\pi$ periodicity will likely apply to any near-DC (as opposed to microwave) measurement. Nonetheless, below we initially try a $4\pi$-periodic candidate CPRs before returning to $2\pi$ periodicity.

Extraction of the Velocity of Dissipative Excitations

Using the phenomenological model of the main text, the velocity $v_{\text{ex}}$ of dissipative excitations in the junction can be extracted from a linear fit of the data in Fig. 2(b) assuming a unity proportionality constant between $I_C R_N$ and $E_C$ (i.e. $I_C R_N=E_C/e$). As $I_C R_N$ is not completely linear in $1/W$, we focus on $1/W < 1 \mu m^{-1}$, yielding $v_{\text{ex}}=1.4\pm0.2\times10^4 m/s$. Electrical transport measurements of the Fermi velocity $v_F$ for surface electrons in Bi$_2$Se$_3$ range from $10^5 m/s$ [S18] to $10^6 m/s$ [S10],
bracketing the value $\nu_F=4.2\times10^5\text{ m/s}$ [S4] extracted from ARPES, and larger than our inferred value. In fact, a lower group velocity is expected for the bound pairs of electrons and holes that shuttle Cooper pairs across the device [S19]. Specifically for Majorana fermions – a subset of these bound pairs – the velocity $\nu_M$ has been predicted to be less than $\nu_F$ by a factor $(\Delta/\mu)^2$ in Ref. [S15] and $(\Delta/\mu)$ in Ref. [S20] (calculated for neutral modes created in graphene JJs), where $\mu$ is the chemical potential of the TI weak link relative to the Dirac point of the surface states. The typical ratio for $\Delta/\mu$ in our samples $\sim10^{-3}$ [S4], giving an estimate for the Majorana velocity of $4.2\text{ m/s for } (\Delta/\mu)^2$ and $4.2\times10^2\text{ m/s for } (\Delta/\mu)$, closer to our measured value though still off by a factor of 20 to 30. The discrepancy between the theoretically predicted value of Ref. [S20] and the velocity extracted from experiment could occur for two reason. First, the proportionality constants in the relationship between $I\!C/R_N$ and $E_C$ and between $\nu_M$ and $\nu_F$ are not known. Second, there is no direct measure of the chemical potential of the surface state in our devices. For samples of density similar to ours, it has been shown that the chemical potential of the surface is less than that of the bulk due to band bending [S4]. This reduction of the surface chemical potential will cause an increase in the expected Majorana velocity.

**Simulation of Josephson Effect in the Presence of Magnetic Flux**

The critical current through the devices as a function of applied field was calculated in a manner closely following that of Tinkham [S5], but with the extended junction model modified to allow non-standard current-phase relationships and spatial inhomogeneities in critical current density. With the flake surface parallel to the $x-y$ plane, applied field $B$ along $z$, and current along $x$, the phase difference between the leads as a function of the position $y$ along the junction is given by

$$\varphi(y) = \varphi_0 + \frac{2\pi}{\Phi_0} \int_0^y \int_0^{L+2\lambda_L} dx B_z(x, y'),$$

(1)

where $\varphi_0$ is the phase difference at $y=0$, $\Phi_0$ is the magnetic flux quantum, $L$ is the length of the junction, and $\lambda_L$ is the London penetration depth. This follows from integrating the vector potential $\mathbf{A}$ around a rectangular contour that includes a path at $y'=0$ and $y'=y$ and taking advantage of the fact that $\mathbf{A}=(\Phi_0/2\pi)\nabla \gamma$ inside the superconducting leads, where $\gamma$ is the non-gauge-invariant phase [defined so that $\varphi = \Delta \gamma - (2\pi/\Phi_0) \int \mathbf{A} \cdot d\mathbf{s}$]. With normalized current-phase relationship (CPR) $i(\varphi) = I(\varphi)/I_C$ and critical current line density $K_c(y)$, we obtain critical current

$$I_C = \max_{\varphi_0} \int_0^W dy K_c(y) i(\varphi(y)).$$

(2)

Note that this analysis is equally valid for $2\pi$- and $4\pi$-periodic phase relations, so long as for the latter case we allow $\varphi(y)$ to range up to $4\pi$ and define $i(\varphi)$ over the full range. For a typical $2\pi$-periodic current-phase relationship $I(\varphi) = I_C \sin(\varphi)$, and uniform current density and field, the critical current drops to zero when an integer number of flux quanta are threaded through the junction area, corresponding to an integer number of cycles of Josephson current. A simulation of magnetic diffraction pattern (MDP) for $i(\varphi) = \sin(\varphi)$ shows the typical Fraunhofer pattern [S5] [Fig. S3(a)]. Deviations from this typical pattern can occur because of non-sinusoidal CPR or from nonuniform critical current distribution along the junction. For a ballistic JJ, the CPR becomes more of a saw-toothed shape, for which the corresponding MDP is shown in Fig. S3(b). There is a small change in the shape of the MDP, but the minima still occur at integer multiples of $\Phi_0$. Aside from a changing CPR, deviations from the Fraunhofer pattern for short junctions can occur for a non-uniform current distribution. Reductions of the flux at first minimum as large as a factor of 2 can occur in the extreme limit when all of the current is concentrated near the edges of the junction [S21]. The MDP in this extreme case is shown for two different current distributions in Fig. S3(c,d). None of these deviations produces a MDP similar to Fig. 3 of the main text.

Using this simulation approach, we empirically attempt to determine a CPR to match the MDP we observe in our devices. Not any CPR is possible: the CPR must be antisymmetric $I(\varphi) = -I(-\varphi)$, it must come from the derivative of the energy of the junction and so must average to zero over a period such that the energy of the junction $E(\varphi) \propto \int d\varphi I(\varphi)$ does not continue to grow as a function of $\varphi$ [S22]. In addition, the CPR must contain a term for the contribution of the bulk, which was estimated in our devices to be smaller than the surface contribution, but not zero. In the following, we assume the bulk has a CPR $I(\varphi) \propto \sin(\varphi)$. To begin, we assume the simplest CPR that is peaked at values of $\pi$ and $3\pi$, with alternating signs but equal amplitude to satisfy the conditions on the CPR. This result is shown in Fig. S4(a), where we see that the inclusion of peaks at $\varphi = \pi, 3\pi$ produces a narrowed MDP. It does not, however, reproduce the additional minima seen in Fig. 3. Note that this CPR is $4\pi$-periodic, as expected for TI JJs with constant Majorana occupancy. The first correction to the simple, peaked CPR above comes from the addition of the bulk term, which we include as a simple sinusoidal dependence on $\varphi$, $\sin(\varphi)$, with 1/5 the amplitude (as estimated above) of the peaks in the CPR (S4B). Comparing to S4B to S4A, additional features in the MDP appear in S4B, are thereby introduced at $n\Phi_0$, where $n$ is an integer. Maintaining only a single strong peak and dip in a $2\pi$ period of CPR cannot produce a minimum in MDP below $\Phi_0$. To get closer to the
FIG. S4: Magnetic diffraction patterns (red) for (a) two gaussian peaks in $I(\varphi)$ at $\varphi = \pm \pi$. (b) the same as (a) but with an added conventional sinusoidal $I(\varphi)$ of 1/5 the amplitude of the anomalous peaks. (c) The same as (a), with additional peaks at $\varphi = \pi/3, \pi/2$ and $4\pi/3, 3\pi/2$, resulting from additional zero-energy crossings. (d) the same as (c) but with a conventional sinusoidal $I(\varphi)$ of 1/5 the amplitude of the anomalous peaks. In each figure, the current-phase relations used to produced the magnetic diffraction pattern – all 4π-periodic – are shown in the inset, and each diffraction pattern is compared to the conventional sinusoidal case (black). (e) (inset) A CPR arising from an ABS spectrum that does not conserve fermionic parity in the junction and contains peaks at $\pi/2, 3\pi/2$. (main) MDP resulting from the CPR of the inset, produces a sub-$\Phi_0$ structure reminiscent of that seen in experiment. (f) same as (e), except with a nonuniform current distribution (inset) used to more closely reproduce the experimental results.

FIG. S5: (a) A 2D plot of the MDP as a function of $\Phi$ and the amplitude of the sinusoidal bulk contribution (Sine Amp). Sub-$\Phi_0$ minima are observed for values of Sine Amp less than $\sim 0.4$. (b) A 1D cut in (a) at a value of Sine Amp=0.5 (red), showing a narrowed central lobe [compared to the Fraunhofer pattern (black)] in the MDP but no minima below $\Phi_0$.

to the CPR from Majorana fermions [S16], a result of difference in Josephson current depending the even versus odd Majorana occupancies in the JJ. The last CPR considered are of the form from Fig. S2(h) – assuming the system always relaxes to the ground state – and are shown in Fig. S4(e,f), where we again have used the multiple zero-energy crossings of Ref. [S17] but now added a nonuniform current distribution that may occur in our junctions due to supercurrent carried by the sides of our device (Note, the sides of the topological insulator are also expected to have a surface state, yet any supercurrent carried by the sides will be unaffected by the applied magnetic field). The MDP of Fig. S4(f) produces minima at values $\Phi_0/4$, $\Phi_0/2$ and $\Phi_0$, near the experimentally observed minima. It is not possible even in principle to determine a unique CPR from the diffraction pattern but can only supply evidence for a peaked CPR. In our simulations, we were unable to obtain sub-$\Phi_0$ dips for any conventional (i.e. non-peaked) CPR, but were only able to obtain MDPs that look similar to our experimental results using peaked CPRs, with multiple peaks over the range $\varphi=0$ to $2\pi$. Even in these cases, we do not get a precise match to the data – we have tried to limit the number of fitting parameters.

MDPs for a varying bulk contribution are shown in Fig. S5. Fig. S5(a) is a 2D plot of the MDP for the CPR of Fig. S4(e) as a function of $\Phi$ and the amplitude of the sinusoidal contribution (Sine Amp). When Sine Amp=0 (all surface), the resulting MDP is the same as in Fig. S4(e), while for Sine Amp=1 (all bulk) the MDP is same as Fig. S3(a). Fig. S5(a) shows that sub-$\Phi_0$ minima
are obtained for values of Sine Amp less than ~0.4. For Sine Amp=0.5 (red), the MDP obtained is shown in Fig. S5(b), where it is seen that a narrowing of the central feature occurs when compared to a Fraunhofer pattern (black), but the minima still occur at integer values of Φ₀. This may account for prior reports finding a relatively conventional MDPs for JJs on TIs. For example, a MDP similar to the one calculated in Fig. S5(b) is seen in Fig. 2(b) of Ref. [S23].

A Graphite Control Device

To confirm that the anomalous features observed in TI JJs are a result of the interface between a TI and a conventional superconductor, a JJ was created using 75 nm-thick graphite as the weak link. The details of the device fabrication are identical to the fabrication of TI JJs, except the Ar ion mill step was not needed to produce low contact resistance. Instead, a 5 min UV-ozone exposure, typical in the creation of graphene devices [S24], was used. The results, taken at T_f=12 mK, are shown in Fig. S6 where it is seen that the value of $I_C R_N$ is much higher at 244 $\mu$V [Fig. S6a], close to the predicted value of 281 $\mu$V and 427 $\mu$V, and $B_C$ is ~13 mT, closer to the predicted value of 14.3 mT (black line) for a device of $(L,W)=(0.05,1)$ μm. The control device allows for the exclusion of two factors that might have caused the anomalous results in TI JJs. In Fig. S6(a), a plot of $V$ vs. $I$ shows a value for the critical current of 8.1 μA on a device of resistance 30.2 Ω. The larger value of $I_C R_N$ obtained for the control device rules out the possibility of the thermal effects caused by poor filtering being a source of the reduced $I_C R_N$ in the TI JJs. In Fig. S6(b), $I_C$ vs. $B$ (red), extracted from a plot of $dV/dI(B,I)$ (inset of S6B) shows a rather conventional dependence on $B$, closely matching the expected pattern (black). The high value of $B_C$ in the control device rules out the possibility of flux focusing [S25] reducing the values of $B_C$ in TI JJs. $B_C$ for the leads is about 13 mT for this device, so we cannot measure more lobes in the MDP.

Supporting References and Notes

S1. A. K. Geim, K. S. Novoselov, *Nature Mater.* 6, 183-191 (2007).
S2. R. M. Potok, Thesis, Harvard Univesity (2006).
S3. J. G. Analytis, *et al.,* *Nature Phys.* 6, 960-964 (2010).
S4. J. G. Analytis, *et al.,* *Phys. Rev. B* 81, 205407 (2010).
S5. M. Tinkham, *Introduction to Superconductivity.* (Dover Publications, Mineola, New York 1996).
S6. K. K. Likharev, *Rev. Mod. Phys.* 51, 101-159 (1979).
S7. D.-X. Qu *et al.,* *Science* 329, 821 (2010).
S8. K. K. Likharev, *Dynamics of Josephson Junctions and Circuits.* (Gordon and Breach Science Publishers, Amsterdam 1986).
S9. G. E. Blonder, M. Tinkham, T. M. Klapwijk, *Phys. Rev. B* 25, 4515-4532 (1982).
S10. B. Sacépé, *et al.,* *Nature Comm.* 2 575, (2011).
S11. A. Furusaki, *Superlat. and Microstruct.* 25, 809-819 (1999).
S12. H.-J. Kwon, K. Sengupta, K., V. M. Yakovenko, *Euro. Phys. J. B* 37, 349-361 (2003).
S13. L. Fu, C. L. Kane, *Phys. Rev. B* 79, 161408 (2009).
S14. C. L. Kane, M. P. A. Fisher, *Phys. Rev. Lett.* 68, 1220-1223 (1992).
S15. L. Fu, C. L. Kane, Phys. Rev. Lett. 100, 096407 (2008).

S16. P. A. Ioselevich, M. V. Feigel’man, Phys. Rev. Lett. 106, 077003 (2011).

S17. K. T. Law, P. A. Lee, Phys. Rev. B 84, 081304 (2011).

S18. M. Veldhorst, et al., arXiv:112.3527 (2011).

S19. A. V. Shytov, P. A. Lee, L. S. Levitov, Phys. Usp. 41, 207-210 (1998).

S20. M. Titov, A. Ossipov, C. W. J. Beenakker, Phys. Rev. B 75, 045417 (2007).

S21. A. Barone, G. Paternò, Physics and Applications of the Josephson Effect. (Wiley-Interscience Publications, Canada 1982).

S22. A. A. Golubov, M. Y. Kupriyanov, E. Il’ichev, Rev. Mod. Phys. 76, 411-469 (2004).

S23. F. Qu, et al., arXiv:112.1683 (2011).

S24. J. R. Williams, L. Dicarlo, C. M. Marcus, Science 317, 638-641 (2007).

S25. M. B. Ketchen, W. J. Gallagher, A. W. Kleinsasser, S. Murphy, J. R. Clem, in Proceedings of SQUID 85 (Berlin 1985) H. D. Hahlbahn, H. Lubbig, Eds., pp. 865.