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Selective area growth and stencil lithography for in situ fabricated quantum devices

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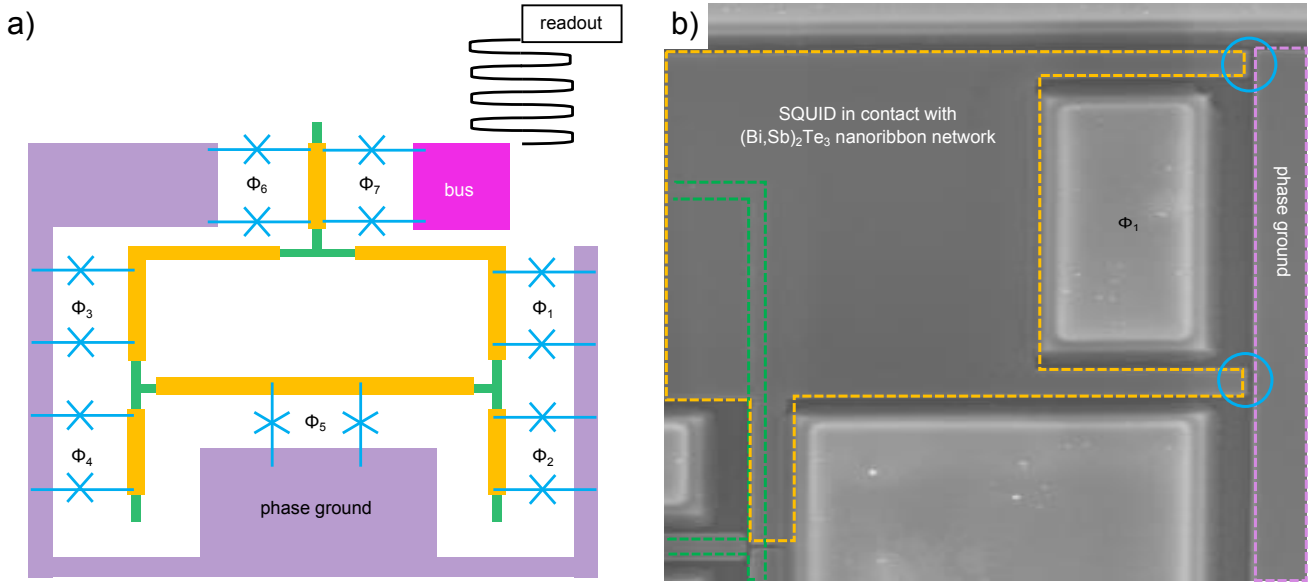
Selective area growth and stencil lithography for *in situ* fabricated quantum devices

-Supplementary Material-

Peter Schüffegen, Daniel Rosenbach, Chuan Li, Tobias W. Schmitt, Michael Schleenvoigt, Abdur R. Jalil, Sarah Schmitt, Jonas Kölzer, Meng Wang, Benjamin Bennemann, Umut Parlak, Lidia Kibkalo, Stefan Trellenkamp, Thomas Grap, Doris Meertens, Martina Luysberg, Gregor Mussler, Erwin Berenschot, Niels Tas, Alexander A. Golubov, Alexander Brinkman, Thomas Schäpers and Detlev Grützmacher

A. Working principle of a Majorana-based qubit.

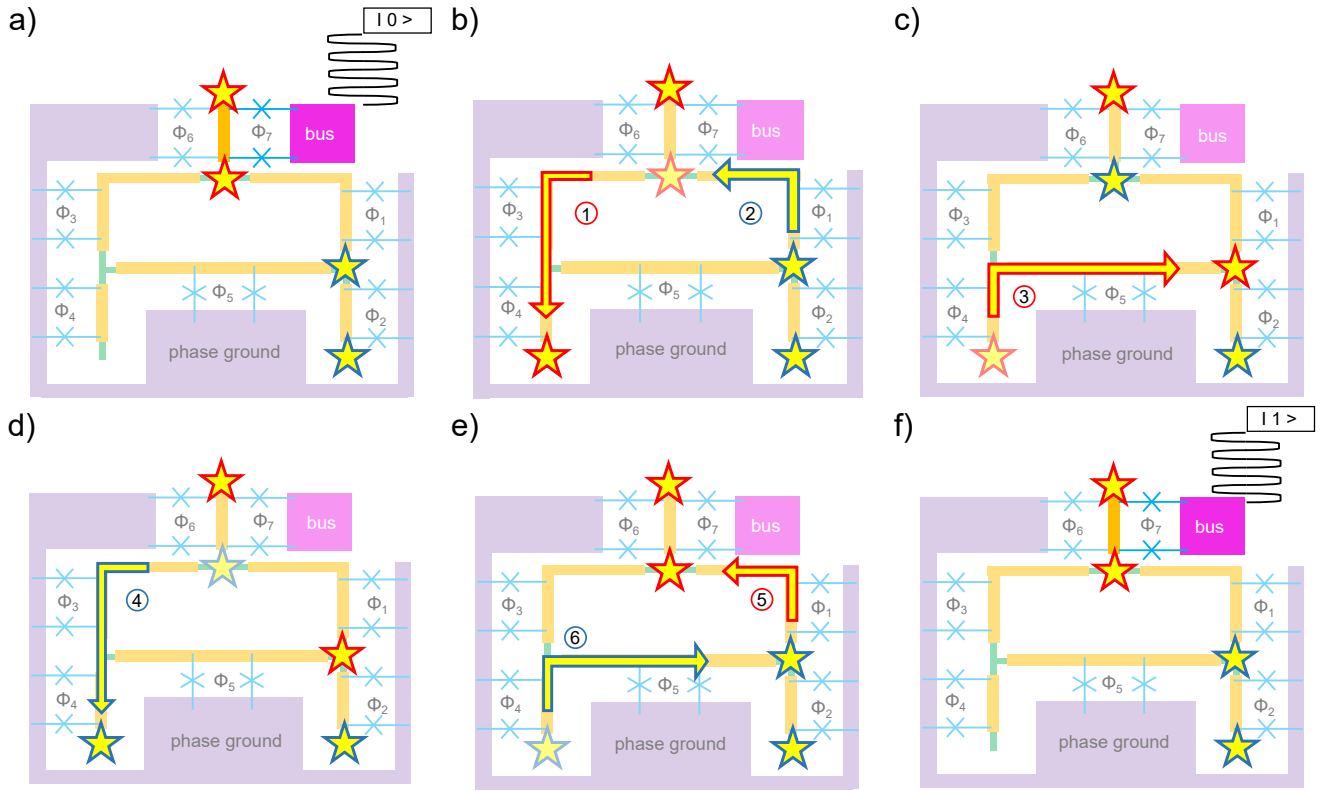
In topologically protected Majorana qubits the information is stored in so-called Majorana zero modes (MZMs), and computations are performed by moving (braiding) these localised modes around each other within a 2-dimensional superconductor – topological insulator (S-TI) network. In order to realise a braiding scheme, either flux-control¹ or electrostatic gates² can be employed. To harness a flux-control, local



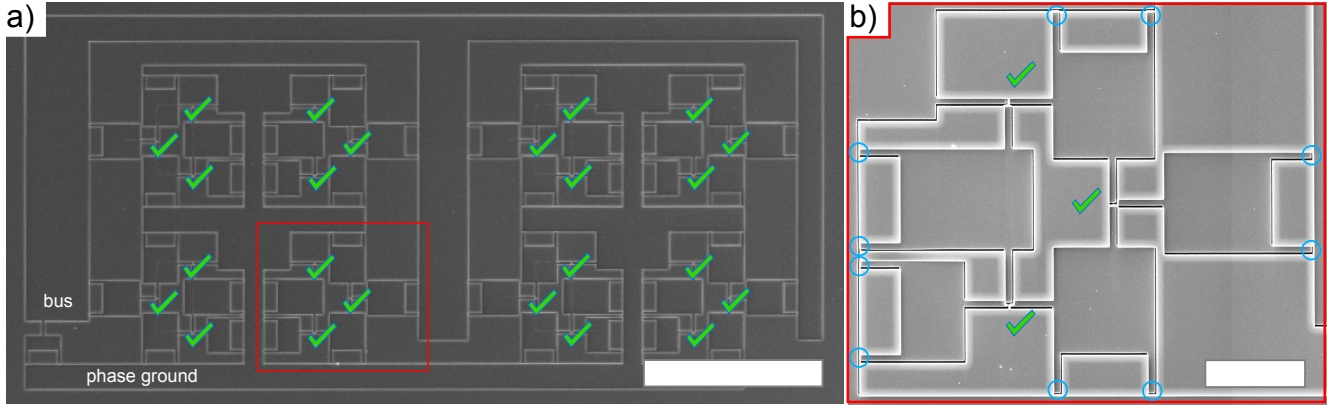
Supplementary Fig. 1: Integration of superconductive periphery (i.e. read-out and flux-control) to the S-TI network for qubit operations. a) A common superconducting phase ground (purple) is connected via two topologically trivial JJs (blue crosses) to each S-island (yellow) of the S-TI network (cf. Fig. 4a main text). A pair of two JJs forms a scanning quantum interference device (SQUID). By changing the local fluxes (Φ_{1-7}) through individual SQUIDs, one can adjust the superconducting phase of the S-islands with respect to the phase ground. By sequences of locally varying fluxes, adjacent S-islands can be (de)coupled, in order to move MZMs within the network (cf. Fig. 2). b) Besides the S-island in contact with the TI-network, one can also deposit the superconducting periphery via the stencil technique. In an additional *ex situ* fabrication step, one can shunt S-islands (yellow frame) and the phase ground (purple frame) by depositing a normal metal (in the areas indicated by the blue circles), in order to define S-N-S junctions. Two S-N-S junctions in parallel thereby form the required SQUID for flux-control.

magnetic fluxes (Φ_{1-7}) need to be applied to well-defined areas on the S-TI chip³ (Fig. 1a). Operating the flux-control via SFQ technologies, which are coupled to the S-TI chip via multi-chip modules, offers a scalable solution⁴. By locally changing the magnetic fluxes Φ_{1-6} in the respective scanning quantum interference devices (SQUIDS) (indicated by a pair of blue crosses in Fig. 1) adjacent S-TI islands (yellow) can be coupled and decoupled. Thereby MZMs move along the TI nanostructure (as shown in Fig. 2). The SQUIDS are defined by the (purple) superconductive phase ground and the S-islands (yellow), which are in contact with the TI network (green).

After *in situ* capping the TI network and polishing of the chip (cf. Fig. 5c main text), the devices are now ready for *ex situ* fabrication of SQUIDS for flux-control. Defining a SQUID can be done by coupling the S-islands of the S-TI network (yellow frame in Fig. 1b) to the phase ground (purple frame) via a metallic weak link (in the area indicated by the blue circle in Fig. 1b) in order to define two trivial S-N-S junctions in parallel, i.e. a SQUID. At this point, we would like to stress that the devices shown here and in the main text resemble mock devices. Besides material specifications (discussed in the main text) in a real topological qubit, the weak links of the SQUIDS would be defined by tunnel junctions in order to reduce



Supplementary Fig. 2: Braiding of MZMs via flux-control: a) It is assumed that the initiated qubit has its two fermionic states (each resembled by the blue or red pair of stars, respectively) unoccupied, defining the ground (or $|0\rangle$) state. b-e) By applying local magnetic fluxes (Φ_{1-6}), MZMs can be moved along the S-TI network, finally exchanging the positions of a blue and a red MZM twice (for details on the flux sequences the reader is referred to the original work of Hyart et al.¹). Exchanging their positions in the same direction twice (either clockwise or counter-clockwise) changes the state of the qubit from $|0\rangle$ to $|1\rangle$ or vice versa. f) By decoupling the red pair of MZMs from the residual S-TI network, one can either read-out its parity or couple it to the bus for multi qubit operations.



Supplementary Fig. 3: Yield of the stencil technique: a) A Random Access Majorana Memory (RAMM) mock device, as proposed by Hyart et al. 1, is comprised of eight topological qubits, which are connected via a bus for multi-qubit operations (scale bar = 50 μm). b) Close-up of red frame in a), showing one qubit integrated into the RAMM. All “normal” bridges (indicated by the blue circles) and all T-shaped bridges (indicated by the green checkmarks) on the chip are intact after *in situ* TI and S deposition (scale bar = 5 μm).

quasiparticles in such circuitry to a minimum. Furthermore, the dimensions of the superconducting islands would differ from those of the theoretical proposals in order to attain a sufficiently large charging energy of the S-TI islands⁵. The layout was adjusted to the theoretical proposals of ref. [1] in order to demonstrate the true-to-scale implementation of the developed *in situ* technology.

In the mock device (Fig. 1b), the phase ground was realised via the stencil technique during the *in situ* deposition of S. For a real qubit, the layout of the stencil mask can be adjusted such that the phase ground is not yet deposited *in situ*. After removal of the stencil mask, a weak link and the phase ground can be defined. In order to implement tunnel junctions instead of SNS junctions a standard tri-layer process can be employed⁶. A five-layer process would even allow for heterogeneous integration of flux control above the SQUIDS for applying the required local magnetic fields for qubit operations.

Once the flux control has successfully been implemented, one should be able to move MZMs within the network by applying magnetic field sequences to the six SQUIDS. It is important to mention that MZMs (indicated as stars in Fig. 2) always come in pairs and that each pair encodes a single fermionic state^{7,8}. This means that the delocalized fermionic state offered by two highly entangled MZMs, can either be occupied by a single electron (here defined as the $|1\rangle$ eigenstate of the topological qubit) or left unoccupied ($|0\rangle$ eigenstate). Superpositions of being empty or occupied are possible as well, resembling the quantum nature of the delocalized state. For measuring whether or not the fermionic state is occupied or unoccupied, one has to read-out the qubit. Depending on the eigenstate into which the qubit collapses during read-out, the resonance-frequency of the transmission line resonator (black line connected to read-out in Fig. 1a), which is used for parity read-out, will differ, providing direct information about the measured eigenstate¹.

The state of the topological qubit can be changed from $|0\rangle$ to $|1\rangle$ by braiding two MZMs around each other twice (Fig. 2). It is important to note that these two MZMs do not belong to the same fermionic state. This is why four MZMs are needed to operate a single qubit. The presented qubit architecture is comprised of six superconductive islands in direct contact with the topological network,

giving rise to twelve MZMs, two at each end of a superconductive island. Six of these MZMs annihilate (due to overlap in T-junctions) and two act as ancillae (for details the reader is referred to the original proposal¹), which leaves the required four MZMs for the desired quantum operation. After initiating the qubit into the ground state $|0\rangle$ (Fig 2a), one can move MZMs of each pair along the TI network by applying sequences of local magnetic fluxes Φ_{1-6} to the SQUIDs. By braiding one red and one blue MZM twice, one changes the state of the qubit from $|0\rangle$ to $|1\rangle$ (Fig 2b-f). Once the braiding operations are successfully completed, the final state can be read-out via moving the MZMs of interest onto the measurement island or bus (pink), decouple them from the residual network via magnetic flux control and read-out the fermionic state via the transmission line resonator. By making the network larger, more MZMs can interact with each other, making more complex quantum computations available. The *in situ* technology allows for fabrication of scalable and complex networks of S and TI, while facilitating a high interface quality. In order to demonstrate the reliability and scalability of the process, a mock device of a Random Access Majorana Memory (RAMM) was fabricated as proposed by Hyart et al.¹ (Fig. 3). In order to estimate the yield of the *in situ* process an exemplary network has been investigated after Nb deposition by scanning electron microscopy, before removing the stencil mask. The RAMM consisted of eight topological qubits, with three T-shaped bridges each. All of the 24 bridges were still intact (indicated by the green checkmarks in Fig. 3). Furthermore, all bridges on the chip, which separate the phase ground from the S-TI network (indicated by the blue circle in Fig. 3b), survived the process. This demonstrates that a 100% yield is achievable for the optimum thickness of the top nitride (~ 100 nm). Cracked bridges were observed on chips which had a thinner top nitride mask layer (< 30 nm).

B. Normal Transport

Complementary to the *in situ* defined Josephson junctions, Hall bar devices were fabricated. For comparison, the same composition of the ternary TI $(\text{Bi}_{0.06}\text{Sb}_{0.94})_2\text{Te}_3$, as mentioned in the main text, was deposited. For the fabrication of the Hall bar devices, a selective area approach was used. Therefore, a Si (111) 4''-Wafer ($>2,000 \Omega\cdot\text{cm}$) was first thermally oxidized using a Tempress oxidation furnace to create a 4.6 nm thin SiO_2 film on top of the Si substrate. The oxidation took place at 820°C for 19 minutes in an oxygen environment. After the oxidation, a 25 nm thick LPCVD grown Si_3N_4 film was deposited on top of the SiO_2 using a Centrotherm E1200 R&D furnace. The top Si_3N_4 was patterned using a combination of structure definition by electron beam lithography and structure transfer into the top Si_3N_4 layer by reactive ion etching. The Si_3N_4 mask layer was etched down to the SiO_2 sacrificial layer, which was removed in a 70 s HF dip before transfer to the MBE system. The HF dip not only etched the SiO_2 to reveal the Si nucleation surface for the TI growth but it also applied a hydrogen passivation layer on top of the pristine Si surface. The hydrogen passivation layer protects the Si surface from oxidation during transfer into the MBE chamber. Before the deposition of the TI, the hydrogen passivation layer was removed *in situ* by heating the substrate to 700°C for 10 minutes.

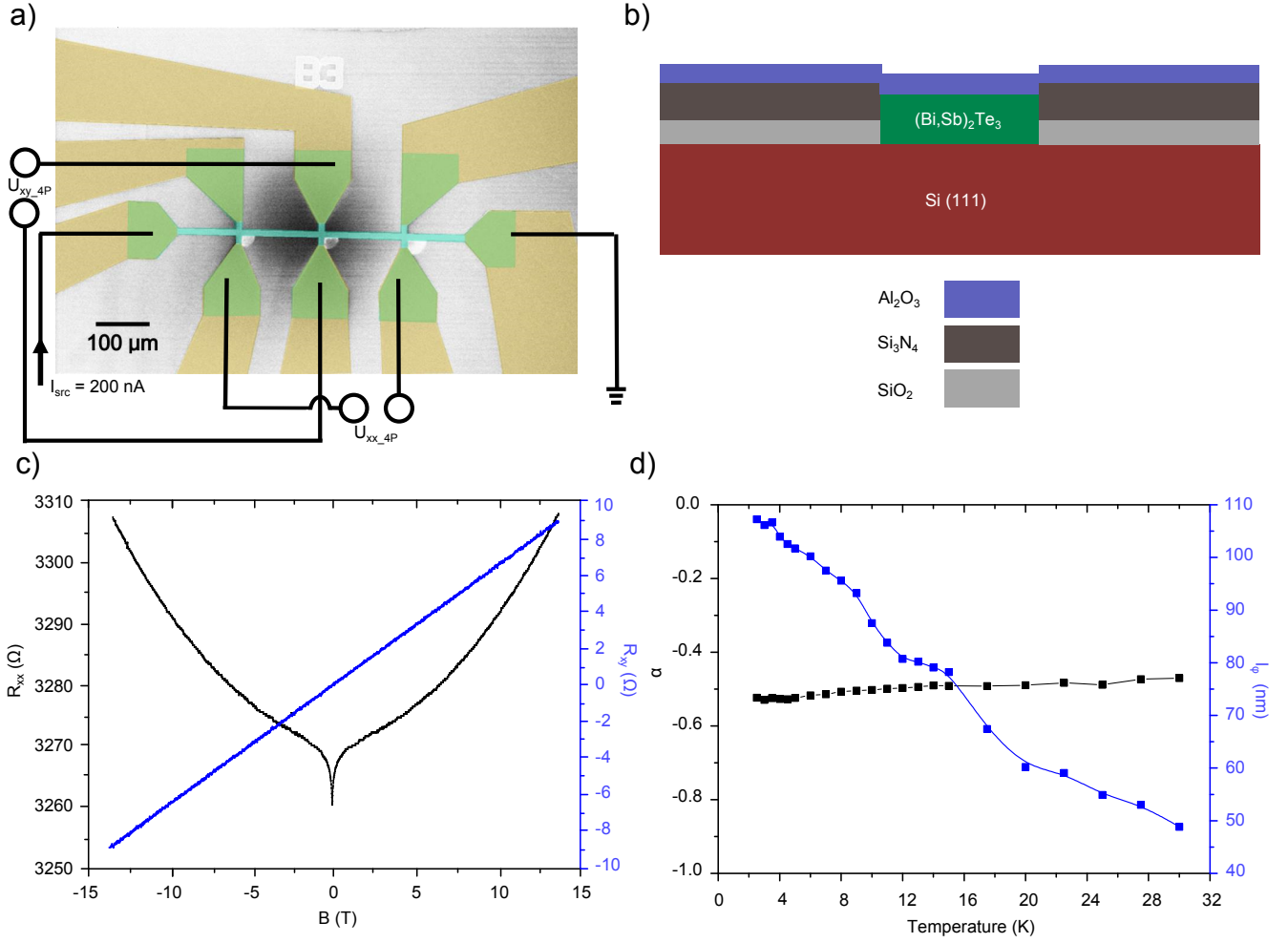
After TI deposition, the substrate was transferred using the portable vacuum chamber. In a second MBE chamber 5 nm stoichiometric Al_2O_3 is deposited on top of the chip covering the selectively grown ternary TI Hall bars without breaking the vacuum. The sample was taken to ambient conditions after the deposition of the dielectric film and electrodes were deposited *ex situ*, using electron beam lithography and metal evaporation of 5 nm Ti and 50 nm Au. Electrodes were defined in the AR-P 6200.09 positive resist. Before metal deposition, the sample was bathed in Fujifilm OPD 4262 developer for 60 s in order to remove the Al_2O_3 passivation layer in the area of the contact pads. Additionally, a gentle Ar^+ -etch was performed prior to metal deposition.

Hall bar measurements on as-prepared devices were performed in a variable temperature insert (VTI) cryostat with a base temperature of 1.5 K and a maximum magnetic field of 13.75 T. The sample geometry resembles a standard eight terminal Hall bar, as can be seen in Fig. 4a). Electrode separation of neighbouring electrodes was measured to be $140 \mu\text{m}$ and the trench width $10 \mu\text{m}$. A schematic of the *in situ* deposited layer stack in the cross section of the Hall bar is shown in Fig. 4b). An AC current of 200 nA amplitude at a frequency of 7.3 Hz was applied in between the outermost electrodes as shown in Fig. 4a). In a four-terminal setup the potential in between two electrodes along the current direction $U_{xx,4P}$ as well as between two electrodes perpendicular to the current direction $U_{xy,4P}$ were measured. The magnetic field perpendicular to the Hall bar surface was then swept from -13.75 T to +13.75 T. The respective transresistance R_{xx} as well as the Hall-resistance R_{xy} as a function of the applied magnetic field is shown in Fig. 4c). The Hall-resistance curve was used to determine the 2D sheet carrier density given by

$$n_{2D} = \frac{1}{q \cdot \frac{dR_{xy}}{dB}} = 9.5 \cdot 10^{14} \text{ cm}^{-2}, \quad (1)$$

where the elementary charge $q = e = -1.62 \cdot 10^{-19} \text{ C}$ and the determined Hall slope were used. The slope of R_{xy} indicates that the majority of the charge carriers are holes. The mobility of the charge carriers was then determined by

$$\mu = \frac{1}{R_{S,B=0} \cdot q \cdot n_{2D}} = 62 \frac{\text{cm}^2}{\text{V} \cdot \text{s}}, \quad (2)$$



Supplementary Fig. 4: Hall bar layout and measurements: a) Hall bar layout of the selectively grown TI Hall bar. The four-terminal measurements setup for the transversal potential $U_{xx,4P}$ as well as the Hall potential $U_{xy,4P}$ is schematically shown. b) Schematic cross section perpendicular to the selectively defined Hall bar. c) Corresponding longitudinal resistance R_{xx} as well as Hall resistance R_{xy} as determined from the Hall measurements performed at 1.5 K. d) Temperature dependent HLN pre-factor α as well as phase coherence length l_ϕ for the measured Hall bar.

with $R_{S,B=0} = 103.57 \, \Omega$ being the sheet resistance at zero magnetic field defined as:

$$R_S = R_{xx} \cdot \frac{W}{L}, \quad (3)$$

The transresistance data was used to determine the phase coherence length by applying the two dimensional Hikami-Larkin-Nagaoka (HLN) formula. The resulting phase coherence length was determined to be $l_\phi = 110$ nm at a base temperature of 1.5 K. The HLN pre-factor was determined $\alpha_{1.5K} = -0.52$ at base temperature, which corresponds to a single transport channel (Fig. 4d). A general explanation for these results is that the top and bottom TI surfaces are coupled via bulk scattering. At first glance this finding seems to contradict the fact that the Fermi level of the upper surface in $(\text{Bi}_{0.06}\text{Sb}_{0.94})_2\text{Te}_3$ can be tuned very accurately into the bulk band gap⁹. However, the discrepancy of normal transport and ARPES in (Bi,Sb)-based TIs can be explained by band bending effects at the upper surface^{10,11}.

Together with the Fermi velocity of $v_F \approx 3.8 \cdot 10^5 \text{ m/s}$ ¹² and the effective mass (m_{eff}) of the hole carriers in pure Sb_2Te_3 ¹³, the elastic mean free path (L_{bulk}) can be derived and the diffusion constant (D_{bulk}) for the bulk of the $(\text{Bi}_{0.06}\text{Sb}_{0.94})_2\text{Te}_3$ thin films:

$$L_{bulk} = \frac{m_{eff} \cdot v_F}{e} = 7.6 \text{ nm}, \quad (4)$$

$$D_{bulk} = \frac{1}{3} L_{bulk} \cdot v_F = 1.4 \cdot 10^{-3} \frac{\text{m}^2}{\text{s}}. \quad (5)$$

Beside bulk states, the Dirac surface states are expected to contribute to the total current. In normal transport, no signatures of the surface channels were found because the normal transport is dominated by bulk states.

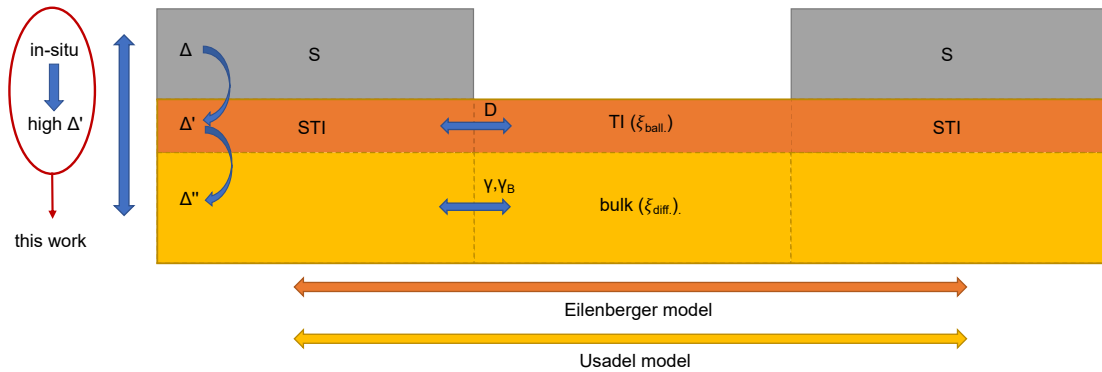
C. Interface Transparency

In total, five *in situ* fabricated Nb-(Bi_{0.06}Sb_{0.94})₂Te₃-Nb Josephson junctions (JJs) were measured in a dilution refrigerator with a base temperature of 12 mK, where the junctions only vary in width (W) and length (L). The normal state resistances across the weak links scale accordingly to the junctions' geometries (see Tab. 1). At low temperatures, a dissipationless supercurrent ($V = 0$) was observed for small DC currents (I) in all junctions (e.g. Fig. 6). Depending on the sweep direction, the retrapping current (I_R), i.e. sweeping I from the dissipative regime towards zero, was much smaller than the switching current (I_c). This hysteresis can be explained by self-heating effects or capacitance¹⁴ and has been reported in topological JJs¹⁵. In Fig. 2e) of the main text the switching current of device 2 as a function of temperature $I_c(T)$ is shown. The slope of $I_c(T)$ reveals a kink at 1.4 K, indicating two contributions to the supercurrent. Recently, a similar observation, which indicated a diffusive and a ballistic channel to a supercurrent in an S-TI-S junction, was reported¹⁶. $I_c(T)$ was fitted for device 2 by quasiclassical superconducting transport equations. We modelled the ballistic component of the surface states with the clean limit Eilenberger equations, using the model by Galaktionov and Zaikin¹⁷. Arbitrary barrier transparencies could be assumed, but no spin-momentum locking was included. For simplicity, it was assumed that the two interfaces have a similar transparency. One of the input parameters is the T_c of the junction (or, equivalently, the value of the proximity gap in the proximitized part of the TI), which can be directly seen from the experimental data to be ~ 4 K (Fig. 2e main text).

From these values, it is possible to deduce the coherence lengths of the ballistic surface modes (ξ_{ball}):

$$\xi_{ball} = \frac{\hbar \cdot v_F}{2\pi \cdot k_B \cdot T_{c,ball}} = 110 \text{ nm}, \quad (6)$$

with k_B being the Boltzmann constant, $\hbar$ the reduced Planck constant and v_F is the Fermi velocity of (Bi_{0.06}Sb_{0.94})₂Te₃ (cf. section B). L over ξ_{ball} ratio of 0.3 serves as another model input parameter (consistent with the estimated clean limit coherence length ξ_{ball} , for the parameters of this junction). We fit the shape of the curve by varying the transparency parameter, and the best fit is obtained with a transparency of $D = 0.95$. The deviation from the full transparency (i.e. $D = 1$), despite the absence of a



Supplementary Fig. 5: Overview of parameters: The proximity induced gap Δ' in the surface states (underneath each electrode) defines the proximitized TI region (STI). This region is separated by a virtual interface (of transparency D) to a region of surface states, which is not covered by any S, defining the non-proximitized surface of the TI. Residual bulk carriers under each electrode also experience a proximity induced pairing potential Δ'' . The interface transparency of the bulk is described by γ and γ_B (see text).

Supplementary Tab. 1: Details of *in situ* fabricated devices 1-5:

device	W [nm]	L [nm]	I_c [μ A]	I_R [μ A]	R_N [Ω]	$I_c R_N$ [μ V]
1	500	54	1.0	0.6	74	74
2	620	30	6.2	2.2	50	310
3	1000	30	14.1	4.6	20	282
4	1000	53	4.0	1.4	43	172
5	480	37	4.0	1.2	53	212

physical barrier, was observed before and could be interpreted from a mismatch in chemical potential¹⁸. Note, that D relates to the in-plane boundary between proximitized TI and non-proximitized TI (Fig. 5). The perpendicular proximity effect from Nb into TI is also strong, as reflected by the large critical temperature of the proximity-induced gap (4 K). This we attribute to the *in situ* deposition of Nb. The number of channels determines the overall scaling parameter that provides a value for I_c . This we used as a free parameter and then deduced from this a number of 40 modes that contribute to the ballistic supercurrent. Taking a ratio of twice the width and the Fermi wavelength as an alternative estimate for the number of modes, one obtains a Fermi wavelength in the surface state of about 31 nm, corresponding to a Fermi energy of about 48 meV away from the Dirac point (assuming linear dispersion).

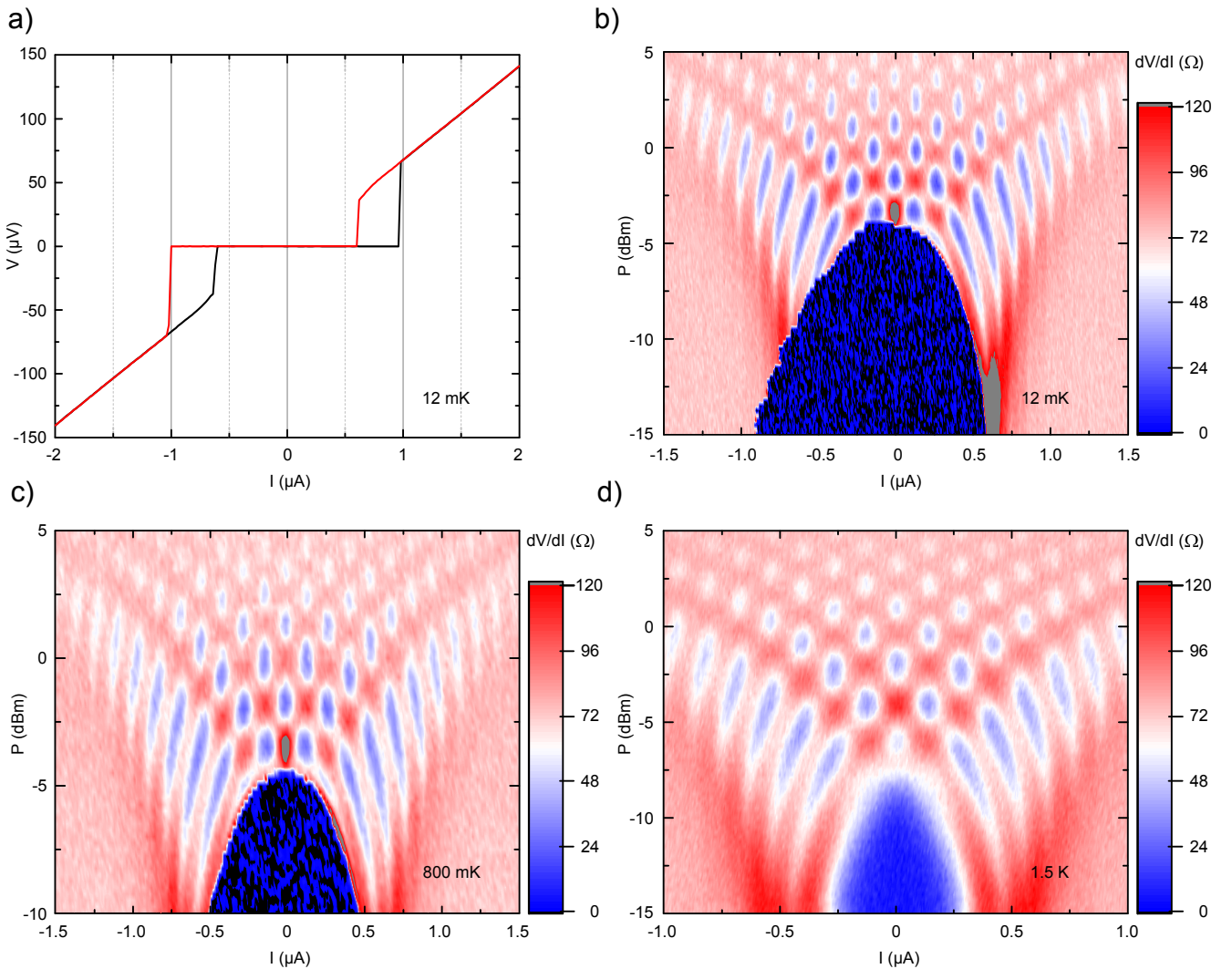
In order to model the diffusive bulk contribution to the Josephson supercurrent the Usadel equations¹⁹ were used with effective boundary conditions²⁰. For junctions with arbitrary length and arbitrary barrier transparency, no analytical expressions exist and a numerical code for fitting the data was used. The interface transparency parameter in this model (γ_B) was set to zero (in practice to $\gamma_B=0.005$ for numerical reasons, but the fitting parameters do not depend on this value). The parameter γ that describes a combination of resistivities across the interface was set to 1. Again, leaving the overall scaling of the current axis free, the only fitting parameters that accurately determine the shape of $I_c(T)$ were then found to be $T_c = 2.0$ K and a length over ξ_{diff} ratio of 2, where ξ_{diff} is the diffusive coherence length:

$$\xi_{diff} = \sqrt{\frac{\hbar \cdot D_{bulk}}{2\pi \cdot k_B \cdot T_{c,diff}}} = 24.6 \text{ nm}, \quad (7)$$

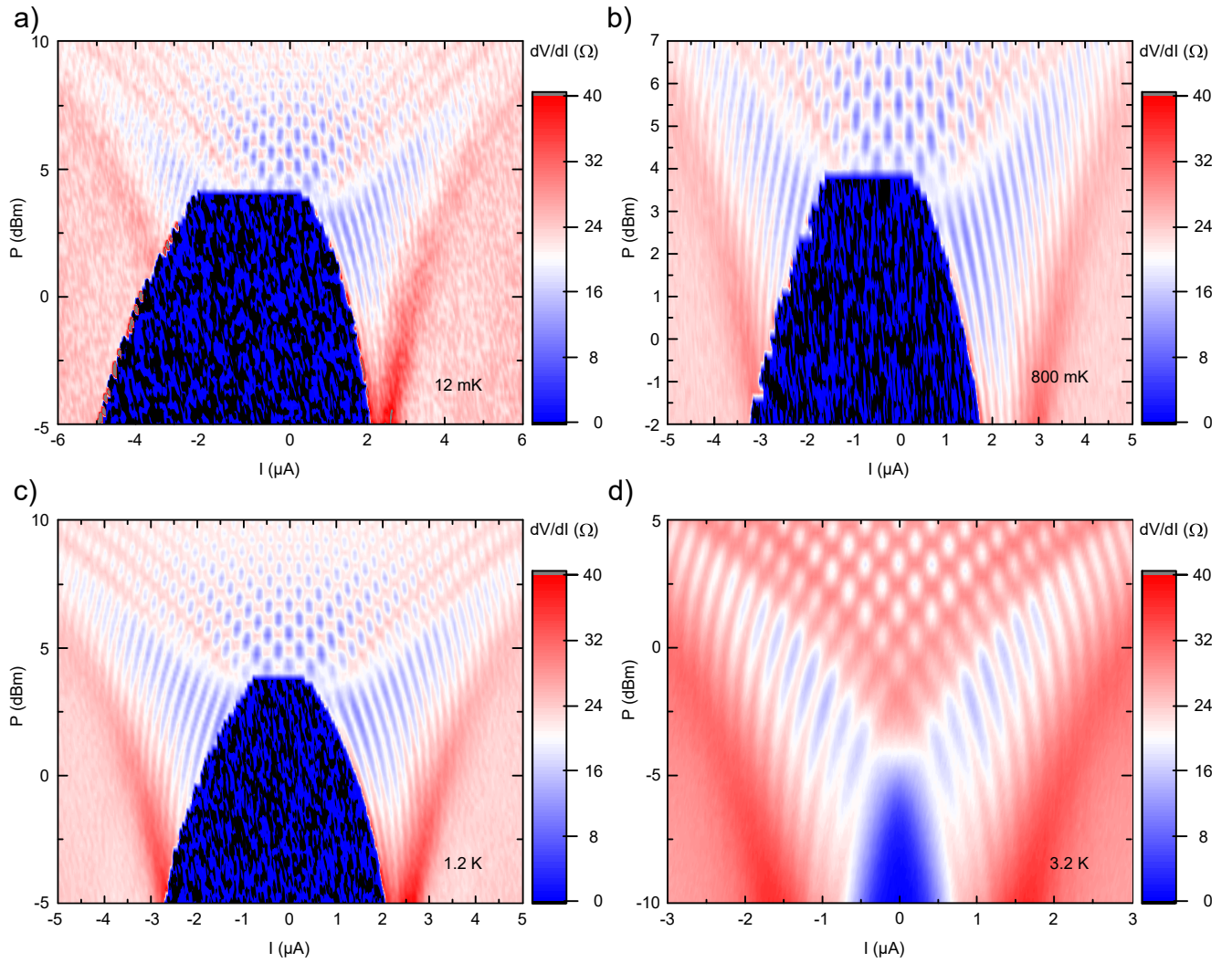
with D_{bulk} being the bulk diffusion constant of $(\text{Bi}_{0.06}\text{Sb}_{0.94})_2\text{Te}_3$, (cf. section B). Its value is consistent with estimates for the electron mobility bias obtained from the Hall bar measurements.

D. Temperature dependent Shapiro response

The I-V sweeps of device 1 are shown in Fig. 6a) for two different sweep directions. Fig. 6b) – d) show differential resistance maps of device 1 for $T = 12$ mK, $T = 800$ mK and $T = 1.5$ K. In Fig. 6b) the differential resistance is shown as a function of I and P (power) of a radio frequency signal with $f_{RF} = 4.6$ GHz at $T = 12$ mK. Under radio frequency irradiation, both the retrapping current (I_R) as well as the switching current (I_C) were slightly reduced. The reduction of the switching current is visible in Fig. 6b), when directly compared to the I – V sweeps in Fig. 6a). In the 2D color plot of the differential resistance the effect of hysteresis is visible. Most of the low-index Shapiro dips at negative currents are covered by the switching current, since I was ramped from $+1.5 \mu\text{A}$ to $-1.5 \mu\text{A}$. In Fig. 6c) and d) the color coded differential resistance is displayed for two elevated temperatures. While at 800 mK a small hysteresis is still present, measurements at 1.5 K do not show any hysteretic behavior. Frequency dependent mea-



Supplementary Fig. 6: Low-temperature measurements of device 1: a) I-V characteristics of device 1 at $T = 12$ mK. The black trace displays the up sweep from $-2 \mu\text{A}$ to $+2 \mu\text{A}$ applied DC bias current. The red trace displays the back sweep (from $+2 \mu\text{A}$ to $-2 \mu\text{A}$) performed on the same junction. b) – d) 2D differential resistance colour plots for an applied radio frequency of $f_{RF} = 4.6$ GHz at three different temperatures as denoted in the graphs.



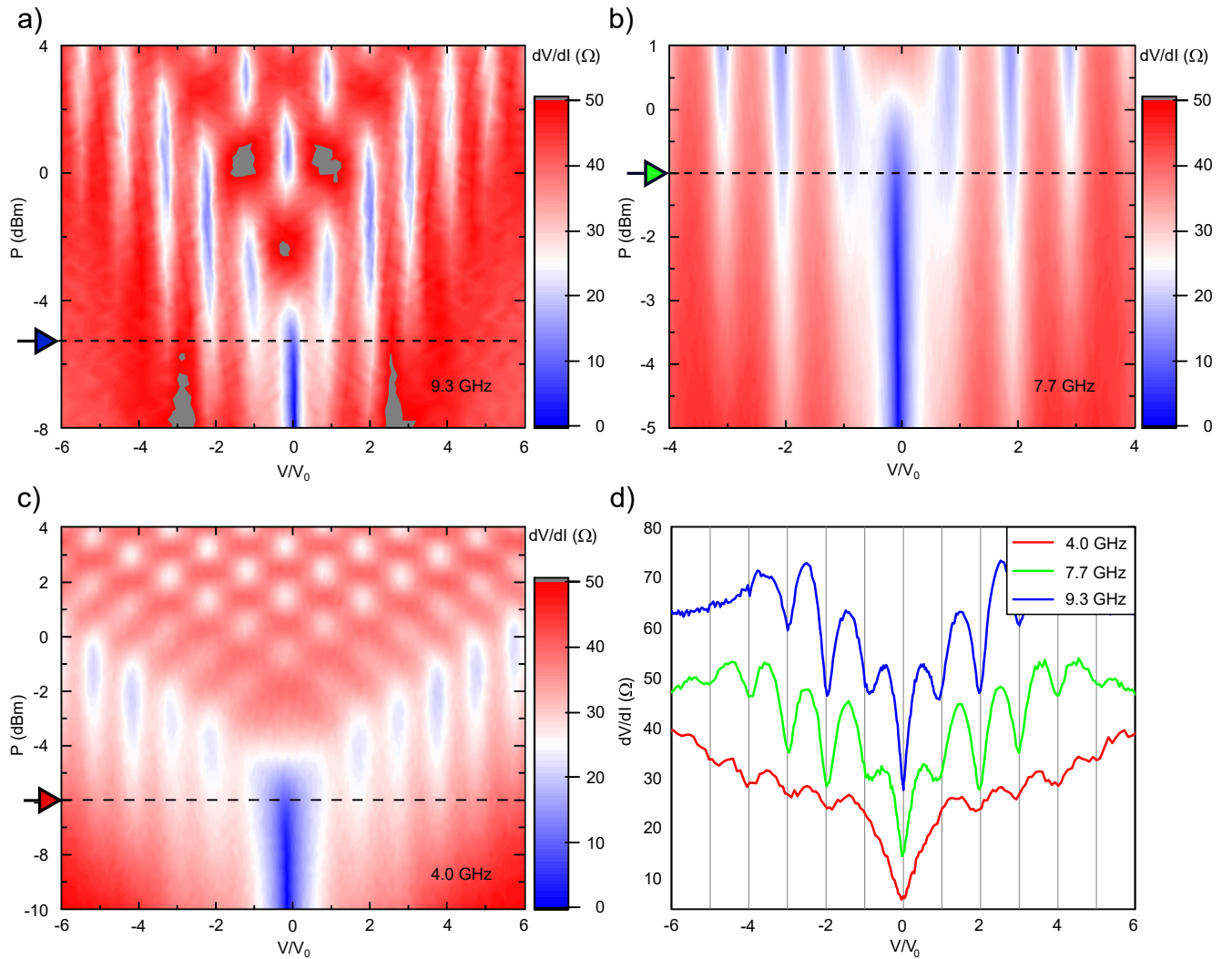
Supplementary Fig. 7: 2D differential resistance colour maps of device 2 for $f_{RF} = 4.0$ GHz: a) $dV/dI(I, P)$ recorded at 12 mK. b) – c) 2D differential resistance colour maps have been recorded at elevated temperatures as denoted in the graphs. d) At 3.2 K no hysteresis is present anymore.

measurements (see Fig. 3 main text) were conducted at 1.5 K to assure that missing dips are not caused by hysteresis.

In addition to device 1, device 2 on the same chip was measured to reproduce the suppression of the first Shapiro dip as a function of frequency. 2D colour plots of the differential resistance are shown in Fig. 7 a)-d). At 12 mK (Fig. 7a) hysteretic behaviour is clearly visible. Even at 1.2 K the hysteresis is still very pronounced, due to the larger switching current (Fig. 7c), and it only vanishes at 3.2 K (Fig. 7d).

E. Shapiro dip renormalization

In order to identify Shapiro dips and to visualize possible missing Shapiro dips, the differential resistance 2D colour plots are normalized to the characteristic Shapiro voltage $V_0 = (h \cdot f_{RF})/2e$. Therefore, in Fig. 3 of the main text the renormalized voltage V/V_0 is shown. The same analysis on the renormalized Shapiro dip data has been performed for device 2 and is shown in Fig. 8a)-d). 2D differential resistance colour plots are shown for 9.3 GHz (Fig. 8a), 7.7 GHz (Fig. 8b) as well as 4.0 GHz (Fig. 8c), each taken at a temperature of 3.2 K. In Fig. 8d) three line scans from the colour plots data in Fig. 8 a)-c) are displayed to compare the evolution of the full integer Shapiro dips for the different radiation frequencies applied. Analogous to the junction presented in the main text (Fig. 3b) at low radiation frequencies (4.0 GHz) the first Shapiro dip is not visible, while it is clearly visible at 9.3 GHz. For 7.7 GHz the first dip is slightly attenuated, which is in accordance with theory²¹ and measurements in HgTe²². The measurements on



Supplementary Fig. 8: Shapiro response of device 2: a) - c) 2D differential resistance colour plots renormalized to the Shapiro step height. Measurements have been performed at 3.2 K for different radio frequencies as indicated in the graphs. d) Line cuts as mentioned in the graphs (A, B, C) are shown for better visibility of the Shapiro dips (differential resistance dips).

device 2 are in line with the results on device 1 (main text), indicating that the suppression of the first Shapiro dip as a function of frequency is a robust and reproducible feature in *in situ* fabricated junctions.

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