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Scanning tunneling microscopy of superconducting topological surface states in Bi_{2}Se_{3}

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In this paper we present scanning tunneling microscopy of a large $\rm Bi_2Se_3$ crystal with superconducting PbBi islands deposited on the surface. Local density of states measurements are consistent with induced superconductivity in the topological surface state with a coherence length of order 540 nm. At energies above the gap the density of states exhibits oscillations due to scattering caused by a nonuniform order parameter. Strikingly, the spectra taken on islands also display similar oscillations along with traces of the Dirac cone, suggesting an inverse topological proximity effect.

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Introduction. Three-dimensional topological insulators (TI) [1], such as Bi_2Se_3 , Bi_2Te_3 or $Bi_{1-x}Sb_x$, were once only known for having great thermoelectric properties. Their most notable physical characteristic, to harbor gapless surface states with striking spin textures protected by topology, was discovered only recently [2–4]. It is understood nowadays that these topological surface states (TSS) stem from a combination of strong spinorbit coupling and band inversion in these materials. It has been quickly realized that studying the interplay of such symmetry-protected states and symmetry-broken phases, including for example magnetism or superconductivity, may lead to a plethora of new effects and provide new platforms for potential technological advances. The most natural approach to achieve this goal is to study a set of phenomena associated with the proximity effect.

It has been predicted that when a topological insulator is brought into contact with a conventional s-wave superconductor (SC) the proximity effect induces superconducting correlations into the TSS that have unconventional p-wave symmetry [5]. This original result triggered a flood of further theoretical works, some representative examples include Refs. [6–11], and a multitude of experimental efforts. The latter span from angleresolved photoemission spectroscopy, scanning tunneling microscopy, point contact, and differential conductance measurements [12–16], through to observations of supercurrents and unusual Josephson Fraunhofer patterns [17– 25]; experiments on phase coherent transport including multiple Andreev reflections, Fabry-Perot interferometry and Aharonov-Bohm oscillations [26–29].

On the technical side, proximity effects can be realized by growing thin layers of SC-TI heterostructures, an approach employed by most of the existing experiments. Alternatively, one could study induced superconductivity in the surface state from the TI's bulk which becomes superconducting by Cu-intercalation into Bi_2Se_3 [30, 31]. While $Cu_xBi_2Se_3$ retains the Dirac surface state, its su-

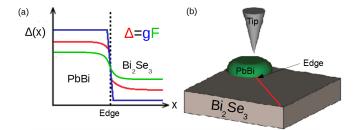


FIG. 1: (Color online) (a) A schematic of the standard proximity effect picture showing the induced superconducting energy gap[34], Δ , at an interface as a function of position x, where F is the superconducting condensate amplitude. The pairing interaction constant, g, is generally taken to have the form of a clear step function, but at small scales, the step actually has a finite slope due to electronic screening [35]. (b) A schematic illustration for the geometry of the STM probe scanning over the surface of TI Bi₂Se₃ in proximity to a PbBi island. In this experiment, we are interested in measuring the superconducting coherence length in the plane parallel to the TI surface, not in the normal direction to the surface as done in many other experiments.

perconducting volume fraction is relatively low which causes obvious challenges. Here we explore a complimentary but different route inducing superconductivity into TSS locally by depositing a matrix of superconducting PbBi islands on the surface of $\rm Bi_2Se_3$. This approach was theorized earlier as a path to creating superconducting graphene [32]. It was employed experimentally to create a tunable realization of two-dimensional superconductivity in mesoscopic superconductor-normal-superconductor arrays [33].

Through the use of cryogenic scanning tunneling microscopy (STM), an experimental technique that is ideal for probing the superconducting proximity effect in inhomogeneous samples, we are able to measure the induced local electronic density of states in TI surface states. A schematic illustration of the experiment is shown in

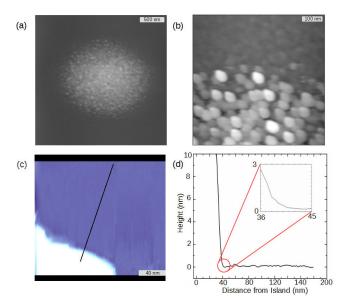


FIG. 2: (Color online) (a) and (b) Atomic force microscopy (AFM) topographs of a thermally deposited PbBi dot. We can clearly see a very grainy appearance to the overall dot, but upon closer inspection it is clear that the dot is comprised of many small superconducting droplets. (c) STM topograph of a PbBi droplet with its respective height profile trace shown in (d). The radius of curvature at the base of the droplet is an artifact due to the radius of the STM tip. The actual interface between the droplet and the TI is very sharp, which is ideal for our measurement.

Fig. 1(b), where an STM probe is placed near the interface of a superconducting island and a TI, and spectra are taken while gradually moving the probe away from the island along the TI surface.

We report in this letter the observation of two main experimental results. First, we show that STM spectra reveal a clear superconducting gap induced into the TSS. From the spatially resolved probes and fits of the gap function we estimate the superconducting coherence length to be of the order of $\lesssim 540$ nm along the direction parallel to the quintuple layers. In addition, at energies above the gap we observe oscillatory behavior of the density of states that resembles the Tomasch interference effect [36]. Second, while all the existing efforts were concentrated on revealing signatures of superconductivity induced into the TSS, much less attention was paid to the corresponding reverse effect of the TSS on an adjacent superconductor. We address this intriguing question by taking careful STM density of states spectra on superconducting islands and uncovering traces of the Dirac cone that seemingly leaks from the TSS. This observation manifestly provides evidence for the possibility of an inverse topological proximity effect.

Measurements and results. The TI used in this experiment is Bismuth doped Bi_{2.04}Se_{2.96}, grown by slowly cooling a stoichiometric mixture of Bi and Se from a tem-

perature of 850 °C. Five atomic planes with atomic order Se1-Bi1-Se2-Bi1-Se1 form a quintuple layer (QL); the QLs are weakly bound to each other, making it possible to readily expose a pristine surface for study. The exposed QL supports the existence of the TSS, which features a single Dirac cone. While $\rm Bi_2Se_3$ is typically n type, the bulk doping of Bi tends to shift the Fermi level back to the center of the band gap [37]. $\rm Pb_{0.3}Bi_{0.7}$ is used as the superconductor due to its favorable whetting properties on $\rm Bi_2Se_3$, its large gap width (3.65 meV), and its high transition temperature of 8.2 K [38].

The TI is first cleaved in a nitrogen environment then placed into a thermal evaporator. Here, we deposit 10 nm of $\mathrm{Pb_{0.3}Bi_{0.7}}$ onto the surface of the TI by evaporation through a TEM mask. This results in large array of superconducting islands with a diameter of $1.2~\mu\mathrm{m}$. From there, the sample is moved to our cryogenic Besocke design STM system for measurement. All of these steps are taken in a vacuum, nitrogen, or helium environment, so the sample is never exposed to air. All STM topographs are taken with a bias voltage of 5 V and tunneling current of 500 pA, and all spectra are taken over a range of -45 meV to 45 meV and measured via a lock-in amplifier.

When doing a surface probe measurement, it is important to fully characterize the surface being measured. In this case, we are interested in the quality of deposition of the PbBi islands as well as how clean the interface is between the island edge and the TI surface. In Fig. 2, we show various atomic force microscopy (AFM) and STM topographs demonstrating the structure of these islands after thermal evaporation; we want to stress that these samples were also not exposed to the air. The PbBi dots appear to be comprised of many 20-100 nm radius droplets grown on top of and around one another. This can clearly be seen in Fig. 2(a) and (b). Fig. 2(c) is an STM topograph showing the edge of one of such droplet formations, along with the respective height profile in Fig. 2(d). Here we can see that the interface between the PbBi and the TI surface is very abrupt, giving evidence of minimal leakage of the PbBi onto the TI surface.

We present the local density of states (LDoS) measurements taken via cryogenic STM at a temperature of 4.2K in Fig. 3. The right panel shows a series of differential conductance plots reflecting LDoS of TSS taken at various distances away from the superconducting island. At a distance of 40 nm the superconducting induced gap is roughly 20% smaller than the corresponding gap on an island, while at distance of order 200 nm the gap falls to almost half of its value.[39] At a distance of > 5 μ m, the Dirac cone is the only dominating feature in the LDoS since the local region of the TI is no longer within the range of the superconducting proximity effect. On the data set taken at 40 nm away from the island one sees oscillatory features occurring with the period of 5-10 mV. As seen in the inset of Fig. 3, the magnitude of the Fourier transform shows a clear resonance at 0.11

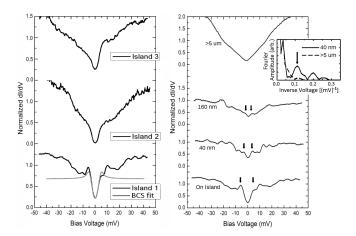


FIG. 3: The right panel represents dI/dV curves measured at 4.2 K taken at various distances from a PbBi island. The LDoS displays clear signature of the induced superconducting gap, with small arrows marking the decoherence peaks. Another notable feature of the presented data is visible oscillations. The inset shows the Fourier transform of the 40 nm data as the solid curve; here the arrow marks the most prominant resonance, corresponding to a periodicity of 9 mV. The left panel represents dI/dV curves measured on different islands at nominally the same conditions. Above the gap one can see not only oscillations but also traces of the Dirac cone at higher energies. All data are normalized so that $dI/dV|_{-20\text{mV}} = 1$. We attribute the asymmetry which can be seen in the left panel to mV-scale offsets between the Fermi energy and the minimum of the Dirac cone, caused by the states leaking onto the superconducting island.

 mV^{-1} , corresponding to a periodicity of 9 mV; a near harmonic is evident at 0.20 mV^{-1} . Interestingly, similar features are present on the LDoS plots taken on the different islands, which are shown on the left panel of Fig. 3. Furthermore, in addition to oscillations at energies above the gap, traces of the Dirac cone are also revealed. We attribute this dependence to the inverse topological proximity effect where LDoS properties of topological surface states penetrate into the superconducting island. In what follows we illustrate this behavior by a simple theoretical model.

Discussion and analysis. It has been emphasized early on that crystals of Bi₂Se₃ possess topological surface states, as well as intra-gap trivial states in the bulk originating from unintentional doping, and importantly also at the surface due to the band bending effect [40]. This coexistence of topological and trivial surface states leads to certain complications in the context of the proximity effect. In particular, phase coherent transport measurements [20, 27, 28] suggest that the superconducting proximity effect of trivial states is in the diffusion dominated transport regime, whereas topological states display transport peculiarities that are specific to the ballistic domain of transport.

In order to gain some theoretical insight into the sys-

tem we have modeled our experimental set-up in two limits.[39] As a first approach we have considered the diffusive limit that should be relevant to a part of the proximity effect governed by the trivial surface states. In complete analogy to previously studied mesoscopic superconductor-normal proximity junctions [41, 42] we solved the standard Usadel equation for a circular geometry describing a superconducting island of radius R surrounded by an infinite normal system. Within this formalism the proximity effect is described by the semiclassical Green's function $G(x,\omega)=\cos[\theta(x,\omega)]$ of position and energy that in the normal region obeys the nonlinear equation

$$\frac{\partial^2 \theta}{\partial x^2} + \frac{1}{x} \frac{\partial \theta}{\partial x} + \frac{i\omega}{E_{\text{Th}}} \sin \theta = 0 \tag{1}$$

where x = r/R is the dimensionless position coordinate and $E_{\rm Th} = v_F l/2R^2$ is the Thouless energy defined by the Fermi momentum v_F and mean free path l. The LDoS is obtained from the real part of the Green's function

$$\nu(x,\omega) = \nu_0 \Re \cos[\theta(x,\omega)],\tag{2}$$

where ν_0 is the normal density of states without the proximity effect. In a linearized regime, applicable at distances away from the boundary where the proximity effect is weak, an analytical result for Eq. (1) is possible,

$$\theta(x,\omega) = \theta_0(\omega) \frac{K_0(x\sqrt{i\omega/E_{\rm Th}})}{K_0(\sqrt{i\omega/E_{\rm Th}})},$$
(3)

elsewhere the problem must be solved numerically. In Eq. (3) $K_0(z)$ is the modified Bessel function and $\theta_0 =$ $\cos^{-1}(\nu_{\rm BCS}/\nu_0)$ and $\nu_{\rm BCS}/\nu_0$ is the BCS density of states on the island. This analysis predicts a reasonable s-wave like proximity induced gap E_g in the normal region, see Fig. 4(a), albeit with a scale set by the Thouless energy, $E_q \sim E_{\rm Th}$, rather than a superconducting gap Δ . However it contains no specific information about the microscopic surface state structure of the material, which is neglected in the semiclassical disordered limit. For the typical known values of $v_F \simeq 5 \times 10^5$ m/s and $l \simeq 80 \text{ nm}$ which are specific to Bi₂Se₃ surface states, and $R \simeq 500$ nm, one estimates a proximity induced Thouless gap $E_g \lesssim 1$ meV to be in a proper parameter regime when compared with Δ . Based on this modeling the expected superconducting coherence length for disordered surface states is in the range of $\xi = \sqrt{v_F l/\Delta} \sim 200$ nm. However, in order to fit the actual spatial profile of the decay of the gap function $\Delta(x)$ in the Bi₂Se₃ found in the experiment [see inset in Fig. 4(b)] it is necessary to use a fitting parameter of the Thouless scale which is different from what is estimated above. The oscillatory features can also not be reproduced in this limit as, although in principle the resulting LDoS in Eq. (2) is oscillating, it follows from Eq. (3) that the oscillation and decay scale

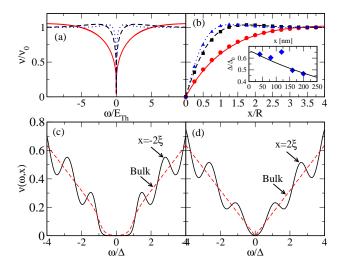


FIG. 4: (Color online) (a,b) The local density of states in the diffusive limit as a function of energy (a) at different distances from the boundary of 0.5R, 1R, 1.5R, and as a function of position (b) at energies $0.1E_{\rm Th}, 0.8E_{\rm Th}, 1.5E_{\rm Th}$, with lines representing the linearized analytical result, and symbols the numerical result of full nonlinear Eq. (1). The inset show a comparison between the gap found experimentally (blue diamonds) and the analytical result (solid line). (c,d) The local density of states as a function of energy for the topological insulator surface states. A phenomenological damping has been included, and the bulk density of states are included as a comparison.

of the Bessel function are controlled by the same parameter.

We then considered the purely ballistic limit of the proximity effect relevant for the topologically nontrivial surface states. Due to technical complications with the underlying equations specific to the spherical geometry, we solved a geometrically simpler planar model. Namely, we have a two dimensional plane consisting of surface states of a three dimensional topological insulator with s-wave pairing in one half of the plain, x < 0. At the mean-field level this is described by Gor'kov's equation:

$$\begin{pmatrix} i\omega_n - H & i\boldsymbol{\sigma}^y \Delta_x \\ -i\boldsymbol{\sigma}^y \Delta_x^{\dagger} & -i\omega_n - H^* \end{pmatrix} \begin{pmatrix} G_{n,k_y}(x,x') \\ F_{n,k_y}^{\dagger}(x,x') \end{pmatrix} = \begin{pmatrix} \delta(x-x') \\ 0 \end{pmatrix},$$
(4)

with a Hamiltonian

$$H^{(*)} = \frac{v_{Fx}}{2} \begin{pmatrix} 0 & \pm \hat{k}_x - ik_y \\ \pm \hat{k}_x + ik_y & 0 \end{pmatrix} + \text{H.c}, \quad (5)$$

which describes the linearized version of the 2D surface states of a 3D topological insulator [4]. $\Delta_x = \Delta\Theta(-x)$ is the s-wave pairing in the superconducting region with Θ the Heaviside theta function as before and $v_{Fx} = v_{FS}\Theta(-x) + v_{FT}\Theta(x)$. $G_{n,k_y}(x,x')$ and $F_{n,k_y}^{\dagger}(x,x')$ are the normal and anomalous Green's functions respectively in the Matsubara representation with fermionic Matsubara frequencies ω_n . Assuming a perfect interface,

the boundary conditions are, $G^T|_{x=0^+} = G^S|_{x=0^-}$ and $F^{\dagger T}|_{x=0^+} = F^{\dagger S}|_{x=0^-}$ and with the proper bulk values of the Green's functions as $x \to \pm \infty$. We have used the superscripts T and S for the topological insulator surface and superconductor regions. The local density of states, integrated over the moment k_y transverse to the boundary, is

$$\nu(x,\omega) = -\frac{1}{\pi} \Im \int_{-\infty}^{+\infty} \frac{\mathrm{d}k_y}{2\pi} \operatorname{tr} \left[G_{n,k_y}^{T,S}(x,x) \right]_{i\omega_n \to \omega + \mathrm{i}\delta} ,$$
(6)

with $G^{T,S}$ taken as appropriate depending on the position of x and the tr performed over spin. By solving the Gor'kov equations one finds for the trace of the Green's function $\operatorname{tr} G^S_{n,k_y}(x,x) = (\omega_n/pv_{FS}^2)(1-e^{-2ipx})$ where $(v_{FS}p)^2 = -[\Delta^2 + \omega_n^2 + (v_{FS}k_y)^2]$. The expression for $\operatorname{tr} G^T$ is similar. The resulting local density of states is then found from Eq. (6) upon an analytical continuation and integration over the transverse momentum

$$\nu(x,\omega) = \frac{|\omega|\Theta(|\omega| - \Delta_x)}{2\pi v_{Fx}^2} \left[1 - J_0 \left(\frac{2x\sqrt{\omega^2 - \Delta_x^2}}{v_{Fx}} \right) \right].$$
(7)

Here J_0 is the Bessel function of the first kind. The bulk density of states in the normal part is $\nu_0(\omega) = |\omega|/2\pi v_{FT}^2$, and in the superconductor we find the bulk density of states $\nu_{S0}(\omega) = |\omega|\Theta(|\omega| - \Delta)/2\pi v_{FS}^2$. Eq. (7) displays Friedel-like oscillations induced in the LDoS in the normal side of the junction. The energy scale of these oscillations, $v_{FT}/2x$, are of the same order of magnitude as those we see experimentally for a typical distance in the range of few hundreds of nanometers. On the superconducting side, Eq. (7) implies oscillatory LDoS with Tomasch-like functional dependence on energy and position, which physically originates from quasiparticle scattering as induced by a nonuniform superconducting order parameter. Even though this modeling was done for a different geometry then that of our system, we believe that these oscillations are generic. Additional analysis shows that for the spherical symmetry Gor'kov equations (4) lead to a LDoS of a more complicated functional form then Eq. (7) with Bessel functions of different harmonics but the essence of the effect remains the same.

Summary. We used superconducting islands deposited on the surface of topological insulator to induce local superconducting correlations into the surface states by virtue of the proximity effect. Superconducting gaps are revealed in the local density of states measurements by cryogenic scanning tunneling microscopy. The gap function is studied as a function of distance away from the island and along the surface state. From these spatially resolved measurements we were able to estimate the range and scale of the proximity effect. We also observed additional oscillatory features in the STM spectra and provided evidence for the signatures of the Dirac cone from the local density of states measurements on super-

conducting islands. Our mesoscopic system will pave the way to further study direct superconducting and inverse topological proximity effects.

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