Evidence for Helical Hinge Zero Modes in an Fe-Based Superconductor

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Abstract

Combining topology and superconductivity provides a powerful tool for investigating fundamental physics as well as a route to fault-tolerant quantum computing. There is mounting evidence that the Fe-Based Superconductor FeTe$_{0.55}$Se$_{0.45}$ (FTS) may also be topologically non-trivial. Should the superconducting order be $s^\pm$, then FTS could be a higher order topological superconductor with Helical Hinge Zero Modes (HHZM). To test the presence of these modes we developed a new method for making normal metal/superconductor junctions via 2D atomic crystal heterostructures. As expected, junctions in contact with the hinge reveal a sharp zero-bias anomaly whose suppression with temperature and magnetic field only along the c-axis are completely consistent with the presence of HHZM. This feature is completely absent when tunneling purely into the c-axis, and its characteristics are also inconsistent with other origins of zero bias anomalies. Furthermore, additional measurements with soft-point contacts in bulk samples with various Fe interstitial contents demonstrate the intrinsic nature of the observed mode. Thus we provide evidence that FTS is indeed a higher order topological superconductor as well as a new method for probing 2D atomic crystals.
Introduction

New particles can be a convincing signature of emergent phases of matter, from spinons in quantum spin liquids to the fermi arcs of Weyl semimetals. Beyond potentially indicating a broken symmetry or topological invariant, they can be put to use in future topological quantum computers. Until recently it was believed the non-trivial topology of the bulk would lead to new states in one lower dimension, that could only exist at the boundary of a bigger system. However, higher order topological insulators (HOTI) have been realized, where the resulting boundary modes exist only at the intersection of two or more edges, producing 1D hinge or 0D bound states. One route to creating these higher order states is through the combination of a topological insulator and a superconductor with anisotropic pairing. Usually this is done by combining two separate materials and inducing superconductivity into the TI via proximity. However, this method requires long coherence lengths and extremely clean interfaces, making experimental realization of devices quite difficult. For studying HOTI, as well as the combination of strong correlations and topology, the material FeTe$_{0.55}$Se$_{0.45}$ (FTS) may be ideal, as it is a bulk, high-temperature superconductor with anisotropic pairing that also hosts topologically non-trivial surface states.
Figure 1: a) Band structure of FeTe$_{0.55}$Se$_{0.45}$ along the Γ-Z and (b) the X-Γ-M cuts. c) Resistance vs. Temperature graph for an exfoliated flake of FTS, showing a clear superconducting transition around 10K. d) Diagram showing the ingredients needed for a Helical Majorana Hinge Mode

FTS is part of the FeTe$_{1-x}$Se$_x$ family of Fe-based superconductors, which ranges from an antiferromagnet in FeTe to a bulk superconductor in FeSe. FeSe has the same Fermiology as the other Fe-based superconductors in that there are hole pockets at the Γ-point and electron pockets at the M-points. The relative strengths of the interband vs intraband scattering in principle should determine the superconducting symmetry, however there is complex interplay between spin-fluctuation exchange, intraband Coulomb repulsion, and the doping level that all contribute to the symmetry of the superconducting order parameter.
While most experiments are consistent with a nodeless $s^\pm$ symmetry, some suggest nodal $s^\pm$, anisotropic $s$-wave, and even $p$-wave. On the theoretical side, different doping levels can lead to $s$-wave, $s^\pm$, $p$-wave, and $d$-wave superconductivity. Nonetheless, experiments performed on FeTe$_{0.55}$Se$_{0.45}$ find no evidence for a node, though strong signatures of $s^\pm$ order. Interestingly, tuning away from FeSe leads to an enhanced spin-orbit coupling and bandwidth. As a result the $p$-orbital is shifted down in energy, crossing the $d$-orbitals with opposite parity along the $\Gamma$ to $Z$ direction (See Figure 1a and b). The first two crossings are protected by crystalline-symmetry resulting in bulk Dirac states above the Fermi energy. However the lowest energy crossing is avoided resulting in a spin-orbit coupled gap, resembling those typically found in topological insulators. While the Fermi level falls into this gap, the original hole and electron Fermi surfaces at $\Gamma$ and $M$, respectively, are retained. ARPES measurements have observed the resulting spin-momentum locked surface states, as well as their gaping out in the superconducting state. Additionally, there is evidence from STM that this results in apparent Majorana zero-modes inside magnetic vortices.

Recent theoretical work on FTS has suggested that the combination of an $s^\pm$ order parameter and topological surface states, could give rise to higher order topological state. In short, the changing superconducting phase causes the surface states to gap out anisotropically. Depending on the relative strength of the isotropic vs. the anisotropic term, this could lead to the [001] and the [100] or [010] face gaping out with opposite phase. As shown in Figure 1d), this is predicted to produce a pair of 1D Helical Majorana Hinge Modes emerging at the 1D interface of the top/side surfaces. Whether or not the modes we observe are indeed Majorana modes, the appearance of HHZM requires both $s^\pm$ superconductivity as well as strong 3D TI surface states. Thus observing Helical Hinge Zero Modes in FTS would provide strong evidence that it is an $s^\pm$ topological superconductor.

To search for the HHZM it is tempting to rely on methods previously exploited to reveal the unconventional nature of the cuprates. Specifically, normal-metal/superconductor junctions demonstrated Andreev Bound States resulting from the $d$-wave order only on [110]
In the case of FTS, this approach is more challenging as one must tunnel into the hinge between [001] and [010] as well as demonstrate the helical nature of the mode. To achieve this, we developed a new method for probing different crystal facets of exfoliated materials. Specifically, we created 2D atomic crystal heterostructures with hBN covering half of the FTS. By draping contacts over the side of the FTS or atop the hBN we can separately probe tunneling into the hinge from the c-axis. As expected for modes protected from back-scattering, we find a cusp like zero-bias peak only on the hinge contacts that is absent from the c-axis junctions. Furthermore its helical nature is revealed by anisotropic suppression in magnetic field. Confirmation that the mode does not result from our fabrication method or defect density is provided by soft-point contact measurements on facets of bulk crystals prepared by different annealing methods (See supplemental Figure S2). Taken together our results strongly suggest the presence of the HHZM in FTS, confirming its higher order topological nature and the presence of $s^{\pm}$ superconductivity.

**Results and Discussion**

The helical hinge zero mode in FTS should be protected from back-scattering only at zero energy as long as the orbital degeneracy is not lifted. Thus, we expect a sharp zero-bias feature on the hinges between the [001] and side surfaces as compared to purely on the [001] face. Alternatively, Majorana zero modes on the hinge should give quantized conductance, revealed through nearly perfect Andreev reflection. However, as discussed later observing this quantized conductance may be challenging as the coherence length in FTS is $\approx 3\text{nm}$. Furthermore, the hinge modes should only exist in the superconducting state and in the absence of magnetic field along the c-axis which breaks the orbital degeneracy. To test this we used 2D atomic crystal heterostructures to simultaneously fabricate Normal Metal/Superconductor (NS) junctions on various crystal facets (See Figure 2a and 2d). The first type of NS junction is a standard lithographically-defined contact that drapes over the
edge of the exfoliated flake. This contact will form a junction with the [001] and [100] surfaces as well as the hinge between them. The second type of contact is fabricated by first transferring hexagonal Boron Nitride (hBN) over half of the FTS flake, insulating the side and edge from electrical contact. We then drape a contact over the side of the hBN, forming a junction primarily on the [001] face (See depiction of the side view in Fig.[2]). From exfoliation to device, the entire process is performed in inert (Ar) or vacuum. Specifically, patterns for mesoscale contacts were defined using standard photolithography techniques and our Heidelberg µPG101 direct-write lithography system. Contact areas are then cleaned with an argon plasma at high vacuum immediately before thermal deposition of 5nm of Cr then 45nm of Au. Full fabrication details can be found in the supplementary.
Figure 2: a) False color image of exfoliated device; numbers denote contacts used. b) $\frac{dI}{dV}$ vs DC Bias voltage for contact 5. c) $\frac{dI}{dV}$ vs DC Bias voltage for contact 3. d) Depiction of contact geometry for top only (5) and hinge (3) contacts. e) Dip number vs. Voltage of dips for c-axis only contacts. The black line is a fit to McMillan-Rowell Oscillations. Blue and red points are experimental data extracted from the positive and negative bias voltages respectively. f) Temperature dependence of differential conductance for various temperatures.

Differential conductance was measured directly with standard lock-in amplifier techniques; the differential conductance vs bias voltage curves for the top only junction and hinge junction are shown in Figures 2b and 2c respectively. As expected from an HHZM, the zero-bias conductance (ZBC) in the hinge contact is quite distinct from the response observed in the top only contact. Specifically, we observe a cusp-like peak in the hinge contact that reaches a zero-bias value 17-times higher than the background or $T \approx T_c$ conductance. Thus providing strong evidence for a zero mode that only exists on the hinge. This size and shape of the response is also completely inconsistent with previous observations of An-
dreev Bound States (ABS), standard Andreev Reflection (AR), Coherent Andreev Reflection (CAR), the Kondo Effect, and Joule heating. The later is immediately eliminated as sweeping the bias voltage back and forth produces no hysteresis in the spectra. In addition, the background conductances in the c-axis, hinge and point contacts are nearly identical, thus heating across all of them should be approximately the same. However, they reveal quite distinct spectra (i.e. strong ZBCP in the hinge contact vs. nearly none in the c-axis), which combined with the emergence of the zero-bias conductance peak (ZBCP) at $T_c$, in numerous contacts (see Figure 2 and supplemental Figure S2), eliminates heating. The ZBCP is also inconsistent with standard AR as the peak is about 17-times the background conductance, whereas AR predicts the peak or plateau to be only twice the background conductance. We also note the normal state resistances of the c-axis and hinge contacts are nearly identical, further eliminating the differences as being due to AR, as this should only depend on the barrier between the contact and the superconductor. The "cusp-like" shape of the peak in combination with the magnitude of the peak suggest ABS, however ABS require either a node in the superconducting gap or time-reversal symmetry breaking, neither of which has been detected. As shown later, ABS, CAR, and the Kondo Effect are all eliminated by the ZBCP response to the direction of applied magnetic field.

Further evidence of the unique nature of our observation results from the temperature dependence of the hinge spectra. Since the HHZM requires both a strong TI and $s^\pm$ superconductivity, we expect its signature to be suppressed alongside the superconductivity. Indeed, as the temperature is increased, the magnitude, but not the width of the ZBCP becomes smaller until the critical temperature, where it is quenched completely. This is in stark contrast to the alternative explanations mentioned above where both the width and the magnitude are reduced with temperature. The spectra also confirm that the contacts draped over the hBN are primarily tunneling in the c-axis. Specifically, above 20 meV we observe a series of dips that are fully consistent with McMillan-Rowell Oscillations (MRO). These MRO result from Fabry-Perot like interference of quasiparticles in the normal layer.
undergoing AR at the interface and reflecting off the back surface of the metal. Indeed, as long established, the MRO are linearly spaced by voltages defined by the metal thickness and fermi velocity (See Figure 2e). The extremely small zero-bias peak seen in this contact likely results either from CAR or the small etching that could expose the hinge. It should be noted that the conductance peak at ±10 meV are not superconducting coherence peaks, but are rather consistent with the spin-orbit coupling gap observed by ARPES (10 meV).

Figure 3: Differential conductance vs bias voltage and magnetic field in the ab-plane (a) or (b) along the c-axis. c) Cut along zero-bias for various applied angles of magnetic field. The cut represents "peak-height", which is the magnitude of the conductance at zero-bias minus the minimum of the spectra. All cuts are normalized to the value at zero-field. d) Polar plot of the percent change of the ZBCP with respect to zero-field for various field strengths.

To test whether the apparent hinge mode is helical, we applied magnetic fields along the different crystallographic directions. As expected, fields along the c-axis strongly suppress the zero-bias peak by breaking the orbital degeneracy between the counter-propagating modes, allowing them to mix and annihilate (See Figure 3). This is in stark contrast to ABS and the Kondo effect where the peak splits in field. To test the importance of orbital versus Zeeman contributions, we measured differential conductance as a function of the angle of the magnetic field with c-axis. As shown in Figure 3, the suppression only depends on the projection of the magnetic field along the c-axis. The fact that it responds to purely orbital and not Zeeman terms further argues against the possibility of magnetic defects causing the ZBCP we observe. While Fe interstitials in FTS are known to point along the c-axis,
we found that our results are insensitive to field versus zero-field cooling. In addition, no hysteresis was observed, let alone apparent switching behavior from in-plane fields. Lastly, as described next, annealing to reduce the Fe interstitials did not reduce the ZBCP.

As a final confirmation that the ZBCP does not result from fabrication, exfoliation, impurities or the specific metal used in the contact, we used the soft-point contact method on bulk crystals. Specifically, bulk crystals were cleaved in inert atmosphere such that numerous terraces were formed. Next, soft-point contacts were made by attaching a copper wire to the crystal via silver paint, which consists of silver nanoparticles suspended in solution. Given the terraced nature of a freshly-cleaved bulk crystal, many contacts are made to various hinges (see inset of Figure 4a). As expected, six different crystals, including one annealed to remove excess Fe interstitials, all reveal spectra nearly identical to the ones observed in the exfoliated device. While the ZBCP tended to be lower, this is as expected given that the contact contains both c-axis and hinge contributions. Furthermore the normal lifetime in the Ag epoxy contact is likely lower, further smearing the spectra and reducing the height at zero bias. Nonetheless, the ZBCP in these soft-point contacts always followed the same temperature and field dependence of the fabricated device discussed above.

![Figure 4: a) Junction Resistance vs. Temperature for soft-point contacts. A superconducting transition happens at 14.5K. Inset is a diagram depicting the contact area of the silver paint. b) Differential conductance vs. Bias Voltage for the junction. c) Differential conductance vs. Bias Voltage as a function of temperature. The ZBCP quenches exactly at the critical temperature of this sample.](image-url)
In summary, we developed a new method to reveal helical hinge zero modes in the topological superconductor FeTe$_{0.55}$Se$_{0.45}$. By contrasting contacts to the [001] surface made using hBN with those draped over the side, we reveal a cusp-like feature in the differential conductance that appears to only exist on the hinge. Further evidence for the helical nature resulted from the suppression with magnetic field pointed only along the c-axis. By combining with measurements using soft-point contacts, we further confirm the intrinsic nature of this new mode. As such, we establish a new means for using 2D-atomic heterostructure devices for probing novel excitations in topological superconductors. Furthermore the appearance of a HHZM in FTS helps to establish both the topological and $s^\pm$ nature of the superconductivity. An important question raised by these results is the large size of the signal coming from the HHZM. It is possible that the large contact size relative to the coherence length at the measured temperature ($\approx 1000x$), makes the measurement essentially many point like contacts in parallel, leading to an apparently large conductance. Thus future theoretical and experimental efforts must be made to better separate out the contact effects from the intrinsic response of the hinge mode we observe.

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