Determination of the charge transfer when tunneling into putative Majorana modes in individual vortices in FeTe$_{0.55}$Se$_{0.45}$

Jian-Feng Ge,$^{1}$ Koen M. Bastiaans,$^{1,2}$ Damianos Chatzopoulos,$^{1}$ Doohee Cho,$^{3}$ Willem O. Tromp,$^{1}$ Tjerk Benschop,$^{1}$ Jiasen Niu,$^{1}$ Genda Gu,$^{4}$ Milan P. Allan$^{1,*}$

$^{1}$Leiden Institute of Physics, Leiden University, 2333 CA Leiden, The Netherlands
$^{2}$Department of Quantum Nanoscience, Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, The Netherlands
$^{3}$Department of Physics, Yonsei University, Seoul 03722, Republic of Korea
$^{4}$Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, NY, 11973, USA

*Corresponding author. Email: allan@physics.leidenuniv.nl

In iron-based superconductors, vortices are reported to host elusive Majorana bound states, but the evidence for their presence remains controversial. Local shot noise measurements have been suggested as a probe into vortex physics, but despite much theoretical work, no local measurement of the shot noise of a vortex core exists. Here, we use local shot noise spectroscopy to study the tunneling process into vortex bound states for the first time, both in the conventional superconductor NbSe$_2$ and in the putative Majorana platform FeTe$_{0.55}$Se$_{0.45}$. We find that tunneling into vortex bound states in both cases exhibits charge transfer of a single electron charge. Our data for the zero-energy bound states in FeTe$_{0.55}$Se$_{0.45}$ excludes the possibility of Yu-Shiba-Rusinov states and is consistent with Majorana bound states. However, it is also consistent with trivial vortex bound states—we argue that further theoretical investigations taking into account dynamics and superconducting tips are necessary.

Introduction

Exotic quantum states and Majorana bound states in vortices

When a type-II superconductor is exposed to magnetic fields, vortices emerge. They are topological line defects where the order parameter vanishes and where quantized magnetic flux penetrates the superconductor.[1] For a clean superconductor with an $s$-wave order parameter, the supercurrent circulating a vortex screens the magnetic field and breaks Cooper pairs into low-energy quasiparticles, which subsequently localize in the vortex core and form a bound state. Vortex bound states reflect both the superconducting and normal-state properties of the underlying superconductor. For example, in the cuprate superconductors, investigations of vortices yielded information on the $d$-wave pairing symmetry$^{[2,3]}$ and peculiar density waves.$^{[4]}$ Yet, in many unconventional superconductors, the nature of the vortex state is still unknown. A very special situation arises in certain iron-based superconductors that are theoretically predicted to be topologically nontrivial. Because of the low Fermi energy $E_F$ relative to the energy gap $\Delta$, a putative zero-energy Majorana bound state would be sufficiently isolated from the trivial Caroli-de-Gennes-Matricon (CdGM) vortex bound states which have an energy spacing of $\Delta^2/E_F$.$^{[5,6]}$ Therefore, Majorana bound states can be experimentally distinguished. Such Majorana bound states embedded in vortices are predicted to obey non-Abelian statistics,$^{[7,8]}$ where braiding the vortices transforms the system from one quantum state to another. It is this nontrivial exchange statistics that underlies the potential importance of Majorana bound states for quantum technology: because these two quantum states encode information nonlocally in a topological qubit with immunity from being corrupted by external...
perturbation, Majorana bound states are predicted to enable fault-tolerant quantum computation.\[^{9,10}\]

So far, the most often reported signature for Majorana bound states is a peak in tunneling differential conductance at zero bias voltage. This signature is readily accessible by experiments, but it is not conclusive proof of the Majorana character of a state.\[^{11–13}\] Other topologically trivial bound states, such as Andreev bound states, can also show the same zero-bias conductance peak, as demonstrated in proximitized superconducting nanowires.\[^{14}\]

Zero-bias conductance peaks in full flux quantum vortex cores\[^{15}\] are also the main evidence for Majorana bound states in the iron-based superconductor FeTe\(^{0.55}\)Se\(^{0.45}\), which is the focus of this study. However, controversy remains as the absence of zero-energy bound states has been reported\[^{16,17}\]. It is still being debated whether the additional observation\[^{18}\]—a saturating conductance at roughly two-thirds of the expected quantized value \(2e^2/h\)—is a strong argument for the Majorana character (\(h\) is the Planck constant and \(e\) is the elementary charge). Such saturating behavior at an arbitrary conductance near \(2e^2/h\) has been observed for Yu-Shiba-Rusinov (YSR) states\[^{19}\] as well; these are present in FeTe\(^{0.55}\)Se\(^{0.45}\) and may also appear as a conductance peak at zero bias.\[^{20}\] Furthermore, it was pointed out that the simple approximations of the Fu-Kane model\[^{21}\] are not likely applicable to the system of vortices in iron-based superconductors,\[^{22}\] which brings the exact nature of the zero-energy vortex bound states in FeTe\(^{0.55}\)Se\(^{0.45}\) into question.

**Shot noise as a probe of electronic matter and to investigate Majorana bound states**

New local probes are thus desired to investigate the electronic properties of the vortex bound states in iron-based superconductors. Recently, it has been pointed out that shot noise, measured at individual vortex cores, could act as a tell-tale probe to distinguish between trivial and Majorana bound states in vortex matter\[^{23}\] and nanowires.\[^{24,25}\] For instance, Ref. [23] suggests shot noise as a differentiator for trivial fermionic states and Majorana bound states. The principle behind these proposals is that Majorana bound states induce resonant Andreev reflection: an incident electron from the coupling lead, when tunneling into a Majorana bound state, is reflected as a hole with unity probability.\[^{26}\] Such a resonant Andreev process is predicted to generate unique Majorana signatures that are absent for trivial fermionic states.

These theoretical studies form the motivation for the shot noise measurements on individual vortex cores that we present here. We measure both the vortex bound states in a conventional superconductor NbSe\(_2\) and the putative Majorana bound states in vortices of FeTe\(^{0.55}\)Se\(^{0.45}\). While we argue that our results do not represent a smoking gun experiment for the existence of Majorana bound states, they allow us to exclude YSR states, and they give a novel experimental insight into these bound states.

Shot noise is, at its core, a consequence of the discreteness of charge. Because of this, tunneling is a Poissonian process, and the noise spectral density \(S\) is proportional to the time-averaged current \(I\),

\[
S = 2q^*|I|.
\]

(1)

Shot noise thus allows probing two quantities that are not visible in the time-averaged current: the effective charge \(q^*\) of the charge carriers and possible correlations between them in electronic matters.\[^{27}\] The former has been used to measure fractional charges in mesoscopic quantum hall systems,\[^{28}\] and the latter has been used to measure the vanishing noise at the quantum conductance of break junctions.\[^{29}\]

Despite a large number of theoretical studies on shot noise tunneling into vortex cores, there has been no experiment so far. The challenge is that one needs high enough sensitivity to
measure the change in $q^*$ from noise, with nanometer resolution to locate individual vortices. This nanoscale resolution is not feasible in mesoscopic setups where noise measurements have been widely applied.

Recently, we have developed scanning tunneling noise microscopy (STNM), which combines scanning tunneling microscopy (STM) and noise spectroscopy, allowing us to measure the effective tunneling charge with atomic resolution.$^{[30]}$ To do so, we build a cryogenic megahertz amplifier that works in parallel with the usual DC measurements, as illustrated in Fig. 1A. STNM has revealed noise centers in cuprates$^{[31]}$ and paired electrons in superconductors.$^{[32,33]}$ STNM also allows to measure shot noise exactly at the core of an individual vortex, which provides a direct and local extraction of the effective charge of the tunneling process into vortex bound states.

**Results**

**Vortex bound states in NbSe$_2$ and FeTe$_{0.55}$Se$_{0.45}$**

In this work, we measure two different materials: the iron-based superconductor FeTe$_{0.55}$Se$_{0.45}$, which is conjectured to host putative Majorana bound states, and the conventional superconductor 2H-NbSe$_2$ as a comparison. We use a tip with an apex made out of Pb, which is a type-I, $s$-wave superconductor with a relatively large gap $\Delta_t \sim 1.3$ meV. We choose to use a superconducting tip in this study for two reasons: first, a superconducting tip provides a superior energy resolution without the limitation from thermal broadening as in the case of a normal-metal tip, i.e. $\sim$0.25 meV (Fig. S1A) instead of $\sim$3.5$k_BT$ = 0.70 meV, where $k_B$ is the Boltzman constant; second, as a consequence of a convolution with the density of states of the superconducting tip,$^{[34,35]}$ the tunneling signal into a zero-energy vortex bound state is effectively shifted from the Fermi level to $\pm\Delta_t$ (illustrated in Fig. S2). This shift circumvents the challenge of measuring shot noise at zero bias voltage.

We first image the subgap electronic structure of the vortices in NbSe$_2$. We introduce vortices by applying an external magnetic field $B = 0.1$ T perpendicular to the sample surface (the upper critical field of the tip $B_c \sim 0.7$ T, see Fig. S1). Because vortices have the strongest enhancement in density of states at the Fermi level of the sample, they are visible as enhanced differential conductance at the energy $|E| = \Delta_t$ when using a superconducting tip (Fig. S2A). Figure 1B shows a spatially resolved image of the differential conductance taken with a sample bias $V_{bias} = -\Delta_t/e$, revealing the full flux quantum ($\hbar/2e$) vortex lattice, with each vortex in the characteristic sixfold star shape for NbSe$_2$.$^{[36,37]}$ We then take differential conductance maps $g(E, r)$ on a fine spatial grid around an individual vortex as shown in Fig. 2. Away from the vortex core, the spectrum in Fig. 2B shows an energy gap with a size of $2(\Delta_t + \Delta_s)$, where $\Delta_s = 1.0$ meV is the superconducting gap of the sample. On the other hand, the spectrum measured at the core center develops two peaks at $\pm\Delta_t$, which translates to a zero-bias conductance peak for a spectrum taken with a normal-metal tip.$^{[38]}$ This translation is confirmed by a deconvolution procedure$^{[20,39]}$ that extracts the local density of states of the sample (see Supplementary Materials for details); as expected, the resulting density of states has a peak at zero energy (Fig. 2E). Deconvolution of spectra along a linecut through the vortex reveals that the zero-bias peak splits away from the core into two dispersing peaks, which eventually merge to the gap edges. These dispersing states are consistent with previous studies$^{[38]}$ of NbSe$_2$ and the expectations of many closely-spaced (on the order of 40 μeV) CdGM bound states from solving the Bogoliubov-de Gennes equations,$^{[40]}$ where the peak at a longer distance from the core center corresponds to a CdGM bound state with a larger angular momentum.
In contrast to the dispersing CdGM bound states in vortex cores of NbSe$_2$, a Majorana bound state is topologically protected such that its energy is locked at the Fermi level. This is exactly what we observe, in agreement with the literature,[15,18,41] in tunneling differential conductance measurements on vortices of FeTe$_{0.55}$Se$_{0.45}$ (Fig. 3). The zero-bias conductance peak does not split (Fig. 3, C and F) as in the case of NbSe$_2$; instead, the non-split bound state extends ~8 nm spatially across the vortex core (Fig. S2), identical to the states observed and interpreted as Majorana bound states in Refs.[15,17,18]. We note that the hybridization between Majorana bound states in a vortex lattice could also split the conductance peaks owing to the spatial overlap of Majorana wavefunctions.[42-44] However, since the average distance between vortices in Fig. 1C is about 120 nm, the energy splitting for the putative Majorana bound states is on the order of 1 μeV.[45]

We then comment on the possibility of the zero-bias conductance peak in FeTe$_{0.55}$Se$_{0.45}$ originating from CdGM bound states as in the case of NbSe$_2$. CdGM bound states should locate not at zero but at a finite energy, and they have been observed—surprisingly only in a subset of vortices in FeTe$_{0.55}$Se$_{0.45}$.[15,16] In these vortices, the lowest energy levels have been reported as small as ~0.1 meV. The question of whether the peak we measured originates from CdGM bound states or not reduces to the accuracy of the peak energy we can determine. From Lorentzian fits (Fig. S2), our results show that the energy of the zero-bias peak is 0 ± 50 μeV, much smaller than the energy of the lowest-lying CdGM bound states reported. Furthermore, a previous high-energy-resolution study[17] shows that the probability of finding vortex bound states having an energy of 0 ± 20 μeV decreases with increasing magnetic field. At a field of 1 T, the chance to find a zero-energy state is 80%. The chance to find a zero-energy state at 0.1 T, the magnetic field we apply in our experiments, is in principle higher. Using 80% as a lower bound, the probability for none of the three vortices we measured that has a state much closer to zero energy than the lowest-lying CdGM bound states is less than 0.8%. In addition, the non-dispersive feature of the vortex bound states we observed in FeTe$_{0.55}$Se$_{0.45}$ is in stark contrast with the dispersive CdGM bound states in NbSe$_2$. Based on our measured electronic structure and the statistics from previous work, it is highly unlikely that the vortex bound states in FeTe$_{0.55}$Se$_{0.45}$ are of CdGM type, in agreement with previous reports.

**Effective charge inside and outside of vortex cores**

The advance of this study is high-sensitivity, atomic-scale noise spectroscopy that allows to extract the effective charge $q^*$ transferred when tunneling into vortex bound states. In the tunneling regime where the transparency is small, we parametrize any changes in noise, including the so-called Fano factor, via the effective charge $q^*$ in Eq. 1 (see Materials and Methods for different definitions of the Fano factor). For example, in the simplest case of electron tunneling, the transferred charge of each tunneling event is a single electron charge ($q^* = 1e$), as expected from Poissonian statistics. In contrast, when Andreev reflection takes place such that an incident electron is reflected as a hole, two electron charges ($q^* = 2e$) are effectively transferred per event. Since our measurements are performed at a finite temperature $T$, the current noise in the tunnel junction with resistance $R$ consists of shot noise and thermal current noise $4k_B T/R$, and takes the form of[27]

$$S = 2q^*(V_{bias}/R)\coth(q^*V_{bias}/2k_BT).$$

This equation, which reduces to Eq. 1 at zero temperature, allows us to extract the effective charge $q^*$ as a function of bias voltage.

We start by measuring current noise at $B = 0.1$ T at locations far away from the vortex cores. There, one expects the noise to correspond to an effective charge of $q^* = 1e$ at bias energies larger than the gap, $|eV_{bias}| \geq (\Delta_t + \Delta_s)$. At these energies, the tunneling of Bogoliubov
quasiparticles dominates the noise. Around the gap energy $\pm(\Delta_t + \Delta_i)$, one then expects a step in noise from $q^* = 1e$ outside the gap, to $q^* = 2e$ inside. This is because, inside the gap, single-electron processes are not allowed anymore, and only Andreev processes contribute to the noise. As shown in Fig 4, our measurements are in qualitative agreement with this picture, both in NbSe$_2$ and FeTe$_{0.55}$Se$_{0.45}$. Outside the gap, our data follows the $q^* = 1e$ line, in agreement with Eq. 2. At the gap energy, a step is visible.

Interestingly, $q^*$ does not reach $2e$ inside the gap, but saturates at a value between $1.3e \sim 1.6e$ (Fig. 4, B and D), despite a vanishing conductance within $\pm(\Delta_t + \Delta_i)$ in Figs. 2B and 3B. This is in contrast to the measurement at $B = 0$ T on FeTe$_{0.55}$Se$_{0.45}$, where the extracted $q^*$ reaches $1.97e$ at $\pm \Delta_i$ (Fig. S6). We hypothetize that the presence of the magnetic field leaves behind a small fraction of delocalized quasiparticles,$^{[46,47]}$ which allows charges of $1e$ to tunnel. Even a very small fraction of quasiparticles will decrease the noise substantially, because for a given tunneling transparency $\tau$, the single-particle processes occur with probability $\tau$, while the Andreev processes occur with probability $\tau^2$ (see Supplementary Materials for estimation of fractions). Future experiments at different magnetic fields, and mapping the exact spatial dependence of current noise around vortex cores are necessary to test this hypothesis relating to the effective charge away from the vortex core. In this study, we focus on the noise spectra in the centers of individual vortex cores.

To investigate the tunneling process into vortex bound states, we then measure the current noise at the vortex cores, first for NbSe$_2$. The experimental data in Fig. 4A show that noise in the core center follows the $1e$-noise behavior until reaching well within $\pm \Delta_i$. We observe a transition from $q^* = 1e$ to $q^* > 1e$ around $\pm 1.0$ meV, within which Andreev reflection at the tip side starts to dominate (Fig. S5A). Nevertheless, the $q^*$ remains at $1.05e$ at $E = \pm \Delta_i$, where tunneling into the vortex bound states occurs (Fig. 4B). As a comparison, we then measured the shot noise of tunneling into the vortex bound states in FeTe$_{0.55}$Se$_{0.45}$. To our surprise, the behaviors of noise and effective charge (Fig. 4, C and D) at the vortex cores of FeTe$_{0.55}$Se$_{0.45}$ are very similar to those of NbSe$_2$, i.e., without any Andreev-reflection enhanced noise at $E = \pm \Delta_i$. We extract an effective charge $q^* = 0.99e$ into the vortex bound states in FeTe$_{0.55}$Se$_{0.45}$, even closer to a single electron charge than that into CdGM bound states in NbSe$_2$.

**Discussion**

**The nature of the zero-energy vortex bound state in FeTe$_{0.55}$Se$_{0.45}$**

We proceed to our discussion of which states are compatible with our results from noise measurements. We start with the possibility that YSR states might cause the zero-bias peak, and shot noise can act as a tell-tale signature here. YSR states originate from the resonant coupling between a superconductor and a magnetic impurity.$^{[48]}$ One can tune the coupling by a local gate, such as a voltage-biased STM tip, and the energy levels of the YSR states shift correspondingly to the local field felt by the impurity.$^{[20]}$ Thus, we expect to see spatially dispersing in-gap conductance peaks when moving the tip away from the impurity. The tunneling process into YSR states, on the other hand, is expected to be dominated by Andreev reflection in the strong tunneling limit (tunneling transparency $\tau > 10^{-3}$).$^{[19]}$ We then expect, in this limit, shot noise with an effective charge of $q^* = 2e$ when tunneling into YSR states.

To confirm this picture, we carry out tunneling conductance and noise measurements on the YSR states in FeTe$_{0.55}$Se$_{0.45}$ with $\tau = 3 \times 10^{-3}$. The YSR states appear as differential conductance peaks with a ring shape around an impurity site (Fig. S6).$^{[20]}$ Our noise measurements when tunneling into these YSR states show enhanced noise and $q^* \approx 2e$ and are indistinguishable from
those of tunneling into the bare superconductor. With their stark contrast to the noise and effective charge of $q^* = 1e$ measured for vortex bound states, we can exclude YSR states as the origin of the zero-bias conductance peak.

**Consistency with Majorana bound states as well as with a trivial origin**

We then turn to the possibility of Majorana bound states as the origin of the zero-bias peak, as put forward by Refs. [15,17,18,41]. Theoretical calculations show that shot noise for tunneling into an isolated Majorana bound state vanishes for Majorana-induced Andreev reflection with unity probability, when the bias energy lies within the width of the Majorana bound state. However, for typical STM measurements where the bias energy is much larger than the intrinsic width of Majorana bound states, the tunneling shot noise is Poissonian, i.e., $q^* = 1e$. On one hand, this indicates that our observation favors the existence of Majorana bound states in vortices of FeTe$_{0.55}$Se$_{0.45}$. On the other hand, our observation of the identical noise behavior and effective charge for CdGM bound states and the putative Majorana bound states also apparently contradicts the theory proposals that shot noise can differentiate between Majorana and trivial bound states. However, we note that the trivial bound state one can distinguish from a Majorana bound state in Ref. [23] is a resonant level in a double-barrier configuration. Tunneling into this kind of resonant level gives shot noise with $q^* = e/2$, which disagrees with our result of $q^* = 1e$ for vortex bound states in FeTe$_{0.55}$Se$_{0.45}$. In addition, no theoretical work has focused on tunneling processes into CdGM states. Thus, more theoretical and experimental studies are needed to understand the tunneling process into CdGM and Majorana bound states in vortices.

**Summary**

We have measured, for the first time, local shot noise when tunneling into vortex bound states in individual vortices of NbSe$_2$ and FeTe$_{0.55}$Se$_{0.45}$. Using a superconducting tip, we demonstrate the feasibility of measuring shot noise even for states close to the Fermi level, which is usually overwhelmed by thermal noise. We demonstrate that the shot noise of vortex bound states in both superconductors appears as Poissonian noise with an effective charge of 1e, in direct contrast with the enhanced noise due to Andreev reflection measured away from vortices or on impurities.

Our results exclude YSR states as the origin of the zero-bias conductance peak at the vortex cores of FeTe$_{0.45}$Se$_{0.55}$. They represent a first step towards determining the exact nature of the state: while they are in agreement with the theoretical prediction for Majorana bound states, we emphasize that we observe an identical shot noise behavior of topologically trivial CdGM bound states in NbSe$_2$. The similarity of shot noise for CdGM and Majorana bound states, as well as its equivalence to that of the single-electron tunneling process, provides a barrier for one to distinguish Majorana bound states from other trivial states by shot noise.

More theoretical work, especially in modeling shot noise with a superconducting tip, might allow to overcome this barrier. We can envision a model for the tunneling process from a superconducting tip into a Majorana bound state, which provides a direct comparison to shot noise with trivial states. Such a model has already been developed but only the tunneling differential conductance was calculated for a temperature much lower than 1 K when using a tip with $\Delta_t \sim 1$ meV. Shot noise, based on this model, would be an interesting direction to provide signatures for Majorana physics. Furthermore, it will be interesting to investigate and understand the departure of the noise from $q^* = 2e$ outside the vortex core using spatially-resolved noise spectroscopy at different magnetic fields.

**Materials and Methods**

6
Different definitions of the Fano factor

While the Fano factor was originally defined as the ratio between the variance and the mean value of a quantity, specific definitions vary in dealing with electrical current and its shot noise. An often applied definition of the Fano factor $F$ is the ratio between the shot noise power $S$ (precisely the Fourier transform of the current-current correlation function) and the Poisson noise $S_P$ due to independent single electrons,$^{[27]}$

$$F = S/S_P = S/2e|I|.$$  

In some theory proposals$^{[24,26]}$ for shot noise of Majorana bound states, one different definition appears where the Fano factor is expressed as

$$F = P/e|I|,$$

where $P$ is the shot noise power (time-averaged current-current correlation function). Another definition of the Fano factor is expressed as the ratio between the differential noise power (the derivative of the time-averaged current-current correlation function with respect to the bias voltage) and the differential conductance,$^{[50]}$

$$F = dP/dV/(e \cdot dI/dV).$$

In the above definitions, however, the transmission of a single electron at a time is assumed. As a consequence, the correlation between them appears as sub- or super-Poissonian shot noise with $F < 1$ or $F > 1$, depending on the details of the transmission probabilities of the conducting channels. In this work, on the other hand, the charge transfer is the quantity of interest, and the STM junction is well in the single-channel, low-transmission regime (our highest tunnel conductance is 0.4 $\mu$S, yielding $\tau < 5.2 \times 10^{-3}$). In this regime, we include the possible correlation between charge carriers in the effective charge, $q^* = S/2|I|$, or, more precisely, following Eq. 2.

Sample preparation and STM measurements

The FeTe$_{0.55}$Se$_{0.45}$ single crystals with a transition temperature $T_C = 14.5$ K were grown using the Bridgman method. The 2H-NbSe$_2$ samples ($T_C = 7.2$ K) are purchased from HQ Graphene. The samples are cleaved in an ultrahigh vacuum at $\sim 30$ K and immediately inserted into a customized STM (USM-1500, Unisoku Co., Ltd). All measurements are performed in a cryogenic vacuum at a base temperature of $T = 2.3$ K. We perform scanning tunneling spectroscopy using standard lock-in techniques. A bias voltage modulation at a frequency of 887 Hz with an amplitude of 100 $\mu$V (for maps around vortex) or 50 $\mu$V (for high-resolution point spectra) is applied. The resulting differential conductance (d$I$/d$V$) values are normalized by setup conductance $I_{set}/V_{set}$. Prior to all the measurements, a Pt-Ir tip is made superconducting by indenting it into a clean Pb(111) surface. Our superconducting tip exhibits a critical field of about 0.7 T,$^{[51]}$ deduced from differential conductance measurements in different magnetic fields on an atomically flat Au(111) surface (see Supplementary Materials for details).

Noise measurements

We perform noise spectroscopy at a constant junction resistance $R_J$ when varying the bias voltage $V_{bias}$ (and hence tunneling current $I = V_{bias}/R_J$) using our custom-built cryogenic megahertz amplifier developed recently. The amplifier consists of an LC circuit and a high-electron-mobility transistor that converts the current fluctuations in the junction into voltage fluctuations across a 50 Ohm line, as described in detail elsewhere.$^{[30]}$ To extract the effective charge transferred in the junction we follow a similar procedure as described in Refs. [32,33].
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Figures

(Fig. 1) Local tunneling shot noise measurements of vortex bound states. (A) Schematic illustration of the STNM setup. If the tunneling process in an STNM experiment is a single electron (gray) into vortex bound states (red), an effective charge $q^* = 1e$ is transferred from the tip to the vortex. When Andreev reflection takes place (blue), a hole (white) is reflected, and the effective charge doubles $q^* = 2e$. HF and LF stand for the high- and low-frequency amplifier, respectively. Full flux quantum $(h/2e)$ vortex lattice in NbSe$_2$ (B) and FeSe$_{0.55}$Te$_{0.45}$ (C) revealed by spatially resolved differential conductance at a magnetic field of 0.1 T. Setup conditions: (B) $V_{set} = -5$ mV, $I_{set} = 200$ pA; (C) $V_{set} = 10$ mV, $I_{set} = 250$ pA.
Fig. 2. Identifying vortex bound states in NbSe$_2$ with a superconducting tip. (A) Spatially resolved differential conductance around an individual vortex at the energy $E = +\Delta_t$. (B) High-resolution differential conductance spectra acquired at the two locations marked by the crosses in (A): the center of the vortex core (red) and off vortex (blue). The red and blue dashed lines indicate the coherence peaks. (C) Differential conductance spectra along the dashed line (purple) in (A) showing the spatial dispersion of the vortex bound states. The gray dashed lines in (B) and (C) indicate the peaks at $\pm \Delta_t$ where tunneling into the vortex bound states occurs at the core center. (D to F) Local density of states (DOS) plots corresponding to (A to C), after deconvolution using the tip DOS. The vortex bound states are indicated by the peak in the local DOS at around zero energy (gray dashed line). Setup conditions: $V_{\text{set}} = 5$ mV, $I_{\text{set}} = 200$ pA.
Fig. 3. Identifying the putative Majorana bound state in FeTe$_{0.55}$Se$_{0.45}$. (A) Spatially resolved differential conductance around an individual vortex at the energy $E = +\Delta_1$. (B) High-resolution differential conductance spectra acquired at the two locations marked by the crosses in (A): the center of the vortex core (red) and off vortex (blue). The red and blue dashed lines indicate the coherence peaks. (C) Differential conductance spectra along the dashed line (purple) in (A) showing the spatial extent of the zero-energy bound state. The gray dashed lines in (B) and (C) indicate the peaks at $\pm \Delta_1$, where tunneling into the putative Majorana bound state occurs. (D to F) Local density of states (DOS) plots corresponding to (A to C), after deconvolution using the tip DOS. The putative Majorana bound state is indicated by the peak in the local DOS at zero energy (gray dashed line). Setup conditions: (A and C) $V_{set} = 10$ mV, $I_{set} = 250$ pA; (B) $V_{set} = 5$ mV, $I_{set} = 250$ pA.
Fig. 4. Local noise spectroscopy on and off vortices in NbSe$_2$ and FeTe$_{0.55}$Se$_{0.45}$. (A and C) Current noise spectra in the tunnel junction ($R_J = 2.5$ MOhm) taken on (red) and off (blue) the vortex shown in Fig. 2A for NbSe$_2$ and Fig. 3A for FeTe$_{0.55}$Se$_{0.45}$, respectively. The locations of these spectra are marked by the crosses in Figs. 2A and 3A with the same colors. Gray curves are the expected noise from Eq. 2 with an effective charge $q^* = 1e$ and $2e$ at $T = 2.3$ K. The dashed lines in (A) and (C) are replicated from Fig. 2B and Fig. 3B, respectively, serving as guides for the coherence peaks (red and blue) and the bound states (gray). The error bars are determined by the fluctuation of the current noise in time, yielding a standard deviation of 9.25 fA$^2$/Hz. (B and D) Effective charge $q^*$ derived by numerically solving Eq. 2 at the energy $E = \pm \Delta_t$ on (red) and off (blue) vortex for three different vortices in NbSe$_2$ and FeTe$_{0.55}$Se$_{0.45}$, respectively.
Supplementary Materials for

**Determination of the charge transfer when tunneling into putative Majorana modes in individual vortices in FeTe\textsubscript{0.55}Se\textsubscript{0.45}**

Jian-Feng Ge,\textsuperscript{1} Koen M. Bastiaans,\textsuperscript{1, 2} Damianos Chatzopoulos,\textsuperscript{1} Doohée Cho,\textsuperscript{3} Willem O. Tromp,\textsuperscript{1} Tjerk Benschop,\textsuperscript{1} Jiasen Niu,\textsuperscript{1} Genda Gu,\textsuperscript{4} Milan P. Allan\textsuperscript{1*}

\textsuperscript{1}Leiden Institute of Physics, Leiden University, 2333 CA Leiden, The Netherlands
\textsuperscript{2}Department of Quantum Nanoscience, Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, The Netherlands
\textsuperscript{3}Department of Physics, Yonsei University, Seoul 03722, Republic of Korea
\textsuperscript{4}Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, NY, 11973, USA

*Corresponding author. Email: allan@physics.leidenuniv.nl

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Fig. S1 to Fig. S8

**I. Characterization of the superconducting tip**

We decorate a mechanically ground Pt-Ir tip with Pb microcrystals by indenting into a Pb(111) single crystal, cleaned beforehand by standard Ar\textsuperscript{+} sputtering cycles, until a superconductor-insulator-superconductor (SIS) tunnel junction is established (Fig. S1A). The energy resolution is estimated to be 0.25 meV from the full width at half maxima of the sharp coherence peaks in Fig. S1A.

Then we perform tunneling spectroscopy with this tip on a clean Au(111) surface at various magnetic fields, as shown in Fig. S1B. The differential conductance measured by tunneling spectroscopy is expressed by

\[
\frac{\text{d}I(V)}{\text{d}V} \propto \int \text{d}E \ N_s(E) \frac{\partial}{\partial V} \{N_t(E + eV)[f(E, T) - f(E + eV, T)]\}, \quad (S1)
\]

where \(E\) is energy, \(N_s (N_t)\) is the density of states (DOS) in the sample (tip), and \(f(E, T) = \frac{1}{[1+\exp(E/k_B T)]}\) is the Fermi function (\(k_B\) being the Boltzmann constant). We model the tip DOS by the Dynes function\textsuperscript{[S1]}

\[
N_t(E, V, \Gamma) = \text{Re} \left[ \frac{E + i\Gamma}{(E + i\Gamma)^2 - \Delta_t^2} \right], \quad (S2)
\]
where $\Delta_t$ is the superconducting energy gap of the tip and $\Gamma$ is the phenomenological broadening parameter (without thermal broadening). The values for $\Delta_t$ and $\Gamma$ are extracted by fitting each spectrum to Eq. S1, assuming a constant $N_s$ for Au(111) in the energy range from -10 meV to +10 meV. The fit results are summarized in Fig. S1C, showing a critical field of 0.7 T, about 7 times larger than that of the bulk Pb.\textsuperscript{S2}

II. Deconvolution of the conductance spectra taken with a superconducting tip

The consequence of using a superconducting tip with an energy gap $\Delta_t$, as illustrated by Fig. S2A, is the resonance tunneling when either of the gap edges of the tip DOS aligns with the zero-energy state (ZES). Thus, the bound state appears as peaks at $\pm \Delta_t$ in the measured differential conductance spectra. Deconvolution of a differential conductance spectrum is necessary to recover the appearance of the bound state at zero energy in the sample DOS, the same as what one would expect for a spectrum taken with a normal-metal tip.\textsuperscript{S3, S4} We follow the deconvolution algorithm described in Refs. [S5, S6] to extract the sample DOS $N_s$ in Eq. S1. We use the fit results at $B = 0.1$ T in Fig. S1C for the tip DOS $N_t$ in the deconvolution.

To determine the energy of the vortex bound states in FeTe$_{0.55}$Se$_{0.45}$, in Fig. S2, B and C we stack the raw and deconvoluted spectra for the line cut in Fig. 3, C and F, respectively. We fit each peak (at $\pm \Delta_t$ for Fig. S2B and at 0 meV for Fig. S2C) with a Lorentzian function, and plot the peak energy as a function of the position along the line in Fig. S2D. We note an additional broadening of the coherence peaks and the zero-bias peak in the deconvoluted DOS (Fig. 3E) compared to the peaks in the raw spectrum in Fig. 3B, due to the need in the deconvolution algorithm to remove oscillatory errors (we chose the optimized value for the control parameter $\gamma = 5.0$ defined in Ref. [S5]). Nevertheless, the peak center locates at 0 ± 50 µeV, confirming them as zero-energy states. On the other hand, from the amplitudes of the Lorentzian fit (Fig. S2E), we find the zero-energy state has roughly an exponential decay in DOS, with a decay length of ~ 4 nm on both sides of the core center.

III. Differential conductance and noise spectroscopy on different vortices

We present the full dataset of both differential conductance and noise spectroscopy performed on all vortices in Figs. S3 (for NbSe$_2$) and S4 (for FeTe$_{0.55}$Se$_{0.45}$), except the ones shown in Figs. 2 to 4. For each material, all vortices exhibit similar behavior as illustrated in the main text, especially a zero-bias peak in the DOS after deconvolution and 1e-noise on vortex. We show in Fig. S5, A and B the extracted effective charge $q^{*}$ as a function of bias energy for the noise spectra in Fig. 4, A and C, respectively. The error bars of $q^{*}$ in Fig. 4B (4D) are extracted from Figs. S5A, S3H, S3P (Figs. 5B, S4H, S4P), within an energy window of 0.2 meV near $\pm \Delta_t$. A bigger error in $q^{*}$ for vortex #2 (and #3) in Fig. 4D is due to a higher junction resistance of 5 MOhm (and 10 MOhm) used for the noise spectroscopy measurements, leading to reduced absolute values of tunnel current and its noise.

IV. Scanning noise spectroscopy at zero field around a YSR impurity

The results of our measurements on FeTe$_{0.55}$Se$_{0.45}$ at zero magnetic field are shown in Fig. S6. Here we observe the Yu-Shiba-Rusinov (YSR) bound states as a ring in the differential conductance map (Fig. S6B). The YSR states lead to a negative differential conductance in the spectrum (Fig. S6D) because of the convolution of a sharp in-gap resonance peak and superconducting tip DOS as we observed in a previous study.\textsuperscript{S6} The noise spectrum measured (Fig. S6E) shows clear transitions from $q^{*} = 1e$ line to $q^{*} = 2e$ line with onsets at $\pm (\Delta_t + \Delta_s)$,
indicating a dominating Andreev reflection inside the gap. This noise behavior is similar to what we observed before on Pb(111) surface with a superconducting tip.\(^{[87]}\) We extract the effective charge \(q^*\) in Fig. S6F by numerically solving Eq. 2. We observe a narrower step of \(q^*\) from \(1e\) outside the gap to 1.99e at \(E = \pm \Delta t\) compared to the broader transition from \(1e\) to a plateau of 1.3e\(-1.6e\) in Fig. S5. The value of \(q^*\) so close to 2e in Fig. S6F indicates the tunneling current originates purely from Andreev reflection in the SIS junction, whereas \(q^*\) short of 2e in Fig. S5 off vortex implies that the tunneling process is not purely Andreev reflection, and that a contribution from \(1e\)-charge tunneling coexists. More importantly, we find that the YSR state does not cause a difference in shot noise, as shown by the noise map in Fig. S6C, compared to the strong ring feature in the differential conductance map taken in the same field of view and at the same bias voltage (Fig. S6B). Therefore, we exclude YSR states as the origin of the zero-energy bound states in the vortex cores of FeTe\(_{0.55}\)Se\(_{0.45}\).

V. The effective charge when single-particle and Andreev processes both contribute

In this section, we calculate, based on an empirical model, the effective charge when both Andreev reflection and quasiparticle of \(1e\) tunneling contribute to the total current. Away from the vortex, the tunnel junction is similar to an SIS junction, as shown by comparing the spectra in Fig. 2B and Fig. 3B and the zero-field spectrum in Fig. S6D. Especially at the bias energy \(E = \pm \Delta t\), the deconvolution yields a vanishing density of states of the sample (Figs. 2E and 3E). Therefore, the tunneling process for this tunnel junction is expected to be dominated by Andreev reflection that transfers a charge of 2e per event. We then introduce a fraction of the \(1e\)-charge tunneling process,\(^{[88]}\) which contributes to current and noise but has no correlation with those of the Andreev process. For a given tunneling transparency \(\tau \ll 1\), the current contributions for the single-particle processes \((I_{1e})\) and the Andreev processes \((I_{2e})\) are proportional to \(\tau\) and \(\tau^2\), respectively,\(^{[89]}\)

\[ I_{1e} \propto n\tau^n/4^{n-1}, \quad n = 1, 2. \]

Here we assume an equal prefactor for both current contributions. The total current is \(I = I_{1e} + I_{2e}\). As \(I_{1e}\) and \(I_{2e}\) are assumed to be independent, the total current noise is the sum of both contributions,

\[ S = 2eI_{1e}\coth(eV/2k_B T) + 2\cdot 2e\cdot I_{2e}\coth(2eV/2k_B T), \]

where the double-charge (2e) transfer is taken into account in the Andreev contribution (the second term). Then we extract numerically the (total) effective charge \(q^*\) by Eq. 2.

Figure S7 plots \(q^*\) as a function of the fraction of \(I_{1e}\) contribution for different junction resistance we used in noise measurements. When \(I_{1e}/I = 0\), i.e., no single-particle process contributes, \(q^* = 2e\) as expected from purely Andreev reflection. Conversely when \(I_{1e}/I = 100\%\), only single-particle process contributes, yielding \(q^* = 1e\). For values of \(I_{1e}/I\) in between 0 and 100\%, we find a quick reduction of \(q^*\) even when a very small fraction of \(I_{1e}\) contribution exists (note the logarithmic scale of the horizontal axis). For example, for \(R_I = 2.5\) MOhm, when \(I_{1e}/I = 0.02\%, \ q^*\) reduces marginally to 1.92e; when \(I_{1e}/I = 3.3\%, \ q^*\) already reduces to 1.07e.

VI. Transparency of the tunnel junction during noise spectroscopy

In differential conductance (Figs. 2 and 3) and noise (Fig. 4) spectroscopy, we use different setup conditions in terms of feedback control of the tip. Specifically, in differential conductance measurements, as the protocols are conventionally applied, feedback is disabled during voltage sweeps (spectroscopy). However, in noise spectroscopy, in order to have
optimal junction stability, we enable a slow feedback to maintain a constant junction resistance (i.e., changing the bias voltage and current setpoint for each point in a sweep), except at the zero-bias point where feedback has to be disabled. As already presented in Fig. 2B, the differential conductance of an SIS junction varies by an order of magnitude during a sweep, the transparency of the junction, if feedback is enabled, could also vary considerably. The transparency $\tau$, which is assumed to be in the $\tau \ll 1$ limit for current noise expressions in the main text, has a significant influence on the resulted noise when it becomes comparable to unity.$^{[89]}$ In Fig. S8A we compare the differential conductance taken with feedback disabled and enabled, at different junction resistance (thus the setup $\tau$). While outside the gap the conductance measured both ways is almost identical, a drastic difference in conductance develops in the gap because of a vanishing quasiparticle density of states. The ratio of the feedback-on conductance over the feedback-off conductance $g_{\text{on}}/g_{\text{off}}$ indicates the enhancement of transparency from $\tau = (R_iG_0)^{-1}$ from an Ohmic current-voltage relation, where $G_0 = 2e^2/h = 77.5$ $\mu$S is the conductance quantum ($h$ being the Planck constant). In fact, the conductance ratio $g_{\text{on}}/g_{\text{off}}$ is barely dependent on $R_i$, as shown in Fig. S8B. To estimate the highest $\tau$ inside the gap throughout our noise measurements around vortices, we approximate $\tau = (R_iG_0)^{-1}$ for the setup bias, where $g_{\text{on}}/g_{\text{off}}$ is close to 1. For simplicity, we model the sample DOS $N_s$ also by the Dynes function (Eq. S2) and calculate the feedback-off conductance $g_{\text{off}}$ by Eq. S1. In this case, the conductance ratio $g_{\text{on}}/g_{\text{off}}$ is equal to the current ratio $I_{\text{on}}/I_{\text{off}}$. Therefore, we first integrate $g_{\text{off}}$ to get $I_{\text{off}}$, and then we obtain the ratio $g_{\text{on}}/g_{\text{off}} = I_{\text{on}}/I_{\text{off}} = VR/I_{\text{off}}$. Using this simple model, we can simulate the ratio $g_{\text{on}}/g_{\text{off}}$ (the green line in Fig. S8B), in good agreement with the experimental results. From Fig. S8 we have confirmed that in our measurement conditions, enabling feedback has a marginal effect on the noise, as the transparency $\tau$ stays below 0.16, which is still in the $\tau \ll 1$ limit.

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Fig. S1. Characterization of the superconducting tip. (A) Differential conductance spectrum after indenting the superconducting tip in a Pb(111) surface. (B) Magnetic-field dependence of the differential conductance spectra (dots) of the tip in (A) on an Au (111) surface. Solid lines show the fit by Eq. S1 to each spectrum. The magnetic field is increased from 0 T (dark red) to 0.7 T (yellow) with a 0.1 T interval. (C) Fit parameters $\Delta t$ (left axis) and $\Gamma$ (right axis) as a function of the magnetic field. The spectra at $B = 0.6$ T and $B = 0.7$ T are almost flat, yielding large error bars of $\Delta t$ and $\Gamma$ in the fit results. Setup conditions: (A) $V_{\text{set}} = 5$ mV, $I_{\text{set}} = 200$ pA; (B) $V_{\text{set}} = 10$ mV, $I_{\text{set}} = 400$ pA.
Fig. S2. Details of the structure of the zero-energy vortex bound states in FeTe$_{0.55}$Se$_{0.45}$. (A) An energy diagram showing the tunneling process from a superconducting tip to the zero-energy state (ZES) in FeTe$_{0.55}$Se$_{0.45}$ with a sample bias $eV_{\text{bias}} = -\Delta_t$. Both the tip and sample are superconducting with a gap size of $\Delta_t$ and $\Delta_s$, respectively. The gray (white) area denotes the occupied (empty) states with a diverging DOS near gap edges. At this bias the diverging quasiparticle DOS leads to a maximal probability tunneling into the ZES (purple), resulting in enhanced differential conductance. (B) Differential conductance and (C) corresponding deconvoluted local DOS spectra for the line cut images shown in Fig. 3, C and F, respectively. Spectra are shifted with a spacing of 0.2 for clarity. A local Lorentzian fit is carried out to each peak inside the gap of each spectrum in (B) and (C). (D) The peak energy and (E) peak amplitude for points at different distances in the line cuts. The peak amplitude is normalized by the maximum of each series: red (blue) for the peak with a positive (negative) energy in (B), and black for the peak near zero energy in (C). The greed lines show a symmetric exponential decay around 10.9 nm (location of the vortex core center) with a decay length of 4.0 nm. Note the logarithmic scale of the vertical axis in (E).
Fig. S3. Differential conductance and noise spectroscopy on different vortices in NbSe$_2$. Differential conductance (A to C, E to G) and shot noise (D) measurements same as shown in Fig. 2 and Fig. 4A, respectively, for vortex #2 in Fig. 4B. (H) The effective charge as a function of bias energy extracted from (D). (I to P) Same as (A to H) for vortex #3 in Fig. 4B.
Fig. S4. Differential conductance and noise spectroscopy on different vortices in FeTe$_{0.55}$Se$_{0.45}$. Differential conductance (A to C, E to G) and shot noise (D) measurements same as shown in Fig. 3 and Fig. 4C, respectively, for vortex #2 in Fig. 4D, except for a junction resistance $R_J = 5$ MOhm used in noise spectroscopy. (H) Effective charge as a function of bias energy extracted from (D). (I to P) Same as (A to H) for vortex #3 in Fig. 4D, except for a junction resistance $R_J = 10$ MOhm used in noise spectroscopy. Note that a YSR impurity is observed, as indicated by the small ring feature next to the vortex in (I).
Fig. S5. Effective charge spectra on and off vortex in NbSe$_2$ and FeTe$_{0.55}$Se$_{0.45}$. (A and B) The effective charge numerically extracted from Fig. 4A and Fig. 4C, respectively, by Eq. 2 in the main text. A further increase of $q^*$ above within ±0.4 meV is caused by an increasing Andreev (and possible multiple Andreev) contribution because of a vanishing tip DOS. Unfortunately, the uncertainty also increases quickly for the numerical solution within this range due to the divergence of the coth function when $V_{bias} \to 0$ in Eq. 2.
Fig. S6. Noise measurements near a YSR impurity at zero magnetic field. (A) STM topography and (B) differential conductance for bias $V_{\text{bias}} = -2$ mV for the same field of view. A ring feature with enhanced (reduced) conductance outside (inside) indicates that resonant tunneling occurs when approaching the impurity. (C) Grid spectroscopic map of noise measured near the impurity [the green square in (B)] at the same bias $V_{\text{bias}} = -2$ mV. The ring feature is absent in noise. (D) Differential conductance spectra measured on and off the impurity, denoted by the red and blue crosses in (B), respectively. (E) Noise spectrum and (F) its corresponding effective charge spectrum taken at off impurity position, showing a step from $1e$- to $2e$-noise. Setup conditions: (A, B, and D) $V_{\text{set}} = 8$ mV, $I_{\text{set}} = 400$ pA; (C) $V_{\text{set}} = -2$ mV, $I_{\text{set}} = 800$ pA; (E) $R_J = 10$ MOhm.
Fig. S7. Simulation of effective charge as a function of quasiparticle current contribution. Three different junction resistance (hence transparency) of 2.5, 5, and 10 MOhm in our experiments are used as an input in the model (see text for details of the model).
Fig. S8. Transparency of the junction with and without feedback control. (A) Differential conductance spectra, normalized by setup conductance, for different junction resistance (solid line, $R_J = 2.5$ MOhm; dotted line, $R_J = 20$ MOhm). Red and blue lines correspond to spectra taken with and without feedback control, respectively. (B) Measured conductance ratio for different junction resistance. The green line shows the simulation result (see text), in good agreement with all the data. The small dip near zero energy originates from the difference between the energy gaps of the tip and sample. Note the logarithmic scale of the vertical axis.