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## Why Shot Noise Does Not Generally Detect Pairing in Mesoscopic Superconducting Tunnel Junctions

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The shot noise in tunneling experiments reflects the Poissonian nature of the tunneling process. The shot-noise power is proportional to both the magnitude of the current and the effective charge of the carrier. Shot-noise spectroscopy thus enables us, in principle, to determine the effective charge  $q$  of the charge carriers of that tunnel. This can be used to detect electron pairing in superconductors: In the normal state, the noise corresponds to single electron tunneling ( $q = 1e$ ), while in the paired state, the noise corresponds to  $q = 2e$ . Here, we use a newly developed amplifier to reveal that in typical mesoscopic superconducting junctions, the shot noise does not reflect the signatures of pairing and instead stays at a level corresponding to  $q = 1e$ . We show that transparency can control the shot noise, and this  $q = 1e$  is due to the large number of tunneling channels with each having very low transparency. Our results indicate that in typical mesoscopic superconducting junctions, one should expect  $q = 1e$  noise and lead to design guidelines for junctions that allow the detection of electron pairing.

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For conventional superconductors, condensation and pairing occur concurrently when cooling below the superconducting critical temperature ( $T_c$ ) [1]. However, for some unconventional [2,3] and disordered [4] superconductors, a state that contains phase incoherent, preformed pairs has been conjectured. Despite a wealth of tantalizing signatures, it is very difficult to distinguish preformed pairs from single electrons because (i) many experimental techniques to measure charge cannot differentiate between two single electrons and one pair, (ii) putative spectroscopic signatures like the pseudogap can have a variety of origins [5–7], and (iii) many experiments are dependent on models to determine whether they indicate paired or single electrons [2]. Yet, to test hypotheses of preformed pairs and bosonic liquids, direct and quantitative experimental information is

needed in order to distinguish between a small fraction of fluctuating pairs and a liquid of bosonic particles.

In principle, shot-noise spectroscopy is such an experimental technique. Because of the discrete nature of the charge carriers, the tunneling process is Poissonian, and thus, the zero-temperature current noise power  $S_I$  of a tunnel junction is proportional to the charge of each charge carrier  $q$ , and the absolute value of current  $|I|$ ,

$$S_I = 2q|I|. \quad (1)$$

Therefore, shot noise can yield information on the effective charge in the system [8]. For a tunnel junction at finite temperature  $T$ , Eq. (1) is modified to  $S_I = 2q|I| \coth(q|V|/2k_B T)$ , where  $V$  is the bias voltage across the tunnel junction, and  $k_B$  is the Boltzmann constant. When the bias voltage is large compared to the temperature ( $qV \gg k_B T$ ), it will reduce to Eq. (1). By measuring shot noise, the charge of the carrier can be directly determined. Shot noise has been used to detect the fractional effective charge in the fractional quantum Hall effect [9,10], pairing in superconductors [11–14], and multiple Andreev reflection in superconducting tunnel junctions [14–16].

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However, in mesoscopic superconducting junctions, with much larger areas where tunneling occurs [as opposed to atomic contacts in break junctions and scanning tunneling microscopes (STMs)], the interpretation of shot noise is not straightforward. For example, noise corresponding to  $q = 1e$  has been measured in Nb/AlO<sub>x</sub>/Nb tunnel junctions, while enhanced noise ( $q > 1e$ ) has been measured for NbN/MgO/NbN tunnel junctions; in both cases, the samples are in the superconducting state [15]. Enhanced noise above  $T_c$  was also measured in a La<sub>2-*x*</sub>Sr<sub>*x*</sub>CuO<sub>4</sub>/La<sub>2</sub>CuO<sub>4</sub>/La<sub>2-*x*</sub>Sr<sub>*x*</sub>CuO<sub>4</sub> mesoscopic tunnel junction [17]. The interpretation of these surprising results is challenging: The noise is much larger than expected and present even outside the superconducting gap. Taken together, it appears that the interpretation of shot-noise experiments in mesoscopic superconducting junctions requires additional consideration. In the present Letter, we present careful measurements of mesoscopic superconducting junctions that yield  $1e$  noise and explain why these do not reveal signatures of pairing.

Before describing our experiments, we discuss the expectation for a typical voltage noise  $S_V$  measurement in a tunnel junction (Fig. 1): It turns out to be very different from the standard V-shaped curve of current noise power  $S_I$  seen in Eq. (1) and reported in current noise measurements in the literature [11,13,14]. The difference stems from the fact that, in practice, the voltage noise  $S_V$  is measured instead of the current noise  $S_I$ . When biased with voltage  $V$ , the voltage noise is given by

$$S_V = S_I R_{\text{diff}}^2, \quad (2)$$

where  $R_{\text{diff}}$  is the differential resistance of the sample. If the  $I$ - $V$  curve is nonlinear, like in a superconducting junction, the shape of the  $S_V(V)$  curve will become complicated. To obtain a quantitative expectation, we can calculate  $S_V$  for a superconductor-insulator-normal-metal (SIN) tunnel junction using the Blonder-Tinkham-Klapwijk (BTK) formulas [14,18–20] [Fig. 1(d); for details, see Supplemental Material [21]]. Our calculation shows that  $S_V$  has a double peak structure inside the gap, the amplitude of which indicates the effective charge; below the superconducting gap, Andreev reflections lead to a doubling of the noise and hence the effective charge from  $q^* = 1e$  to  $q^* = 2e$  [Fig. 1(e)]. Note that this model is for SIN junctions, which feature single Andreev reflections only. In contrast, superconductor-insulator-superconductor (SIS) junctions may give rise to multiple Andreev reflections [22], which further enhance the shot noise inside the superconducting gap at low energies [14–16]. Here we only consider single Andreev reflections.

Currently, there are two widely used state-of-the-art methods for measuring shot noise. The first method measures noise across a wide bandwidth at low frequencies using a room-temperature amplifier [17]. The key

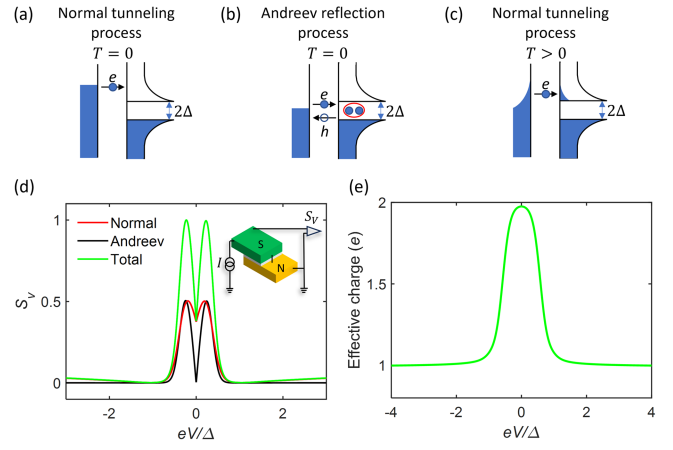


FIG. 1. Tunneling process and shot noise in a NIS tunnel junction. (a) Illustration of single electron tunneling at biases larger than the gap. Black lines indicate the density of states of the superconductor and the normal metal; filled states are shaded in blue. (b) Andreev reflection process when the bias voltage is smaller than the gap, leading to  $2e$ -shot noise. (c) At finite temperatures, normal tunneling processes occur inside the gap. (d) Normalized voltage noise power calculated for different process ( $\Delta = 3.8$  meV,  $T = 4$  K, and transparency  $\tau = 0.01$ ). Red line: noise curve assuming only normal tunneling processes. The finite value at zero bias is thermal noise. Black line: shot noise from Andreev reflection processes. Green line: total noise, which is what is measured in experiments. Inset: schematic of the electrical circuit. (e) Effective charge calculated from the total noise.

advantage of this approach is its broad bandwidth, enabling one to distinguish frequency-independent shot noise from frequency-dependent  $1/f$  noise. The second method measures noise at high frequencies within a narrow bandwidth by employing an  $LCR$  resonator with an impedance matching circuit [13,14,23–25]. Its strength lies in its ability to mitigate the impact of  $1/f$  noise and random telegraph noise due to the higher operating frequency. However, neither of these methods is suitable for the specific experimental challenges presented here. The first method has pronounced  $RC$  roll-off above 100 kHz [17], which makes it impossible to effectively extract shot noise from the total signal in our case. This is because the low-frequency portion of random telegraph noise exhibits a frequency-independent characteristic [26], similar to shot noise, and because the roll-off makes it difficult to measure small signals, considering that the values of capacitance and wire resistance may fluctuate with environmental changes. The second method designed for a system with resistances of the order of  $M\Omega$  or more [13,14,23–25] does not work for the low resistances ( $\sim\Omega$ , due to the finite junction area) found in mesoscopic tunneling junctions as studied here [27]. The issue is that this leads to a quality factor that is too low to use the benefits of an  $LCR$  resonator.

We therefore built a new low-noise, high-frequency amplifier designed to work in the frequency range of

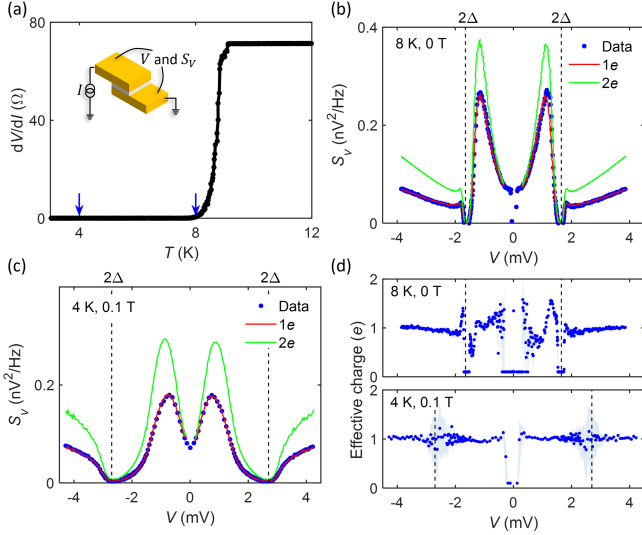


FIG. 2. Shot-noise measurements on SIS junctions. (a) Zero bias  $dV/dI$  from Nb/Al – AIO<sub>x</sub>/Nb sample C1. The blue arrows indicate the temperatures of the noise measurements shown in (b),(c); the inset shows the schematic of the sample. (b) Measured voltage noise data (blue dots) and calculations of the shot noise with  $q = 1e$  (red line) and  $q = 2e$  (green line), at 8 K and zero magnetic field. Black dashed lines indicate the superconducting gap. (c) Noise measurements at 4 K and 0.1 T. (d) Effective charge at different bias voltages. The shaded areas indicate the uncertainty of the data.

100 kHz to 5 MHz, and to fulfill three main requirements: (i) a low-temperature environment to suppress the thermal noise from the amplifier, (ii) a high-resolution amplifier to detect small signals, and (iii) low  $1/f$  noise. Details of the amplifier, including calibration methods and uncertainties of the experimental parameters are given in the Supplemental Material [21].

We start our experiment with a planar Nb/Al – AIO<sub>x</sub>/Nb (SIS) tunnel junction with a zero resistance  $T_c \sim 8$  K (Fig. 2). Tunneling spectroscopy shows the superconducting gap of the Bogoliubov density of states and the resulting nonlinear  $I$ - $V$  curve, which significantly influences our noise curves, as is evident from Eq. (2). We deal with this challenge by first measuring the differential resistance  $R_{\text{diff}}$  of the sample and then calculating the expected shot noise for  $q = 1e$  and  $q = 2e$  according to

$$S_V = 2q|I| \coth(q|V|/(2k_B T)) R_{\text{diff}}^2. \quad (3)$$

These expectations can then be compared with the experimental results or to determine the effective charge from the experimentally measured noise at different bias voltages.

We start with noise data taken at higher temperatures, close to zero resistance  $T_c$ . The temperatures are measured by a calibrated thermometer, and the uncertainty of the temperature measurements is around 0.01 K. Figure 2(b)

shows both the experimental data and the expected noise for  $q = 1e$  and  $q = 2e$  according to Eq. (3). It is obvious that the experimental data overlap very well with the  $q = 1e$  theoretical line both outside and inside the gap. Next, we turn our attention to lower temperatures. Such measurements pose an additional complication due to the appearance of a supercurrent. As the differential resistance becomes zero, the voltage noise also reduces to zero according to Eq. (3) when there is supercurrent in the sample. To solve this problem, we apply an out-of-plane magnetic field of 0.1 T to suppress the supercurrent. The sample is in the center of the magnetic field, and the homogeneity of the magnetic field is 0.1% over a 10 mm diameter spherical volume. With 0.1 T, the supercurrent disappears but quasiparticles are induced as well; for details, see Supplemental Material [21]. From the theoretical calculation, shot noise will still be enhanced inside the gap even if there are more quasiparticles (normal tunneling process). However, we find that the noise still clearly corresponds  $q = 1e$ , as shown in Fig. 2(c). The effective charge stays at  $q = 1e$  within the uncertainty [Fig. 2(d)].

An alternative method to avoid supercurrents is to use a SIN junction. We therefore measure noise at 8, 6, 4, and 2 K in a NbN/oxide/Ag SIN junction with  $T_c \sim 15$  K (Fig. 3). For SIN junctions the zero bias  $dV/dI$  is different from SIS junctions [Fig. 2(a)]. For SIN junctions, when the temperature decreases below  $T_c$ , the gap will develop and the zero bias  $dV/dI$  will increase, while for SIS junctions (Fig. 2), the  $dV/dI$  will become zero due to the appearance of supercurrent. The experimental results at 2 K are shown in Fig. 3(b). Again, the measurements clearly indicate  $q = 1e$  outside and inside the gap, even though the temperature  $T = 2$  K is much lower than  $T_c$ . The noise characteristics at 8, 6, and 4 K are shown in Fig. S5 of the Supplemental Material [21]; they also indicate that  $q = 1e$ .

In Fig. 4, we summarize the effective charge we measured inside the superconducting gap at different

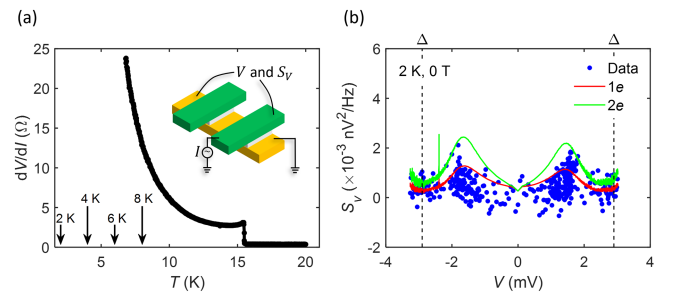


FIG. 3. Noise measurement from a NbN/oxide/Ag (SIN) junction N1. (a) Zero bias  $dV/dI$  measured with constant ac current of 0.25 mA. Inset: top view of the sample. The yellow color indicates NbN; the green colors indicates Ag. Arrows indicate the temperatures at which noise spectra have been measured. Noise data at 2 K without magnetic field are shown in (b); data of other temperatures are shown in Supplemental Material [21].

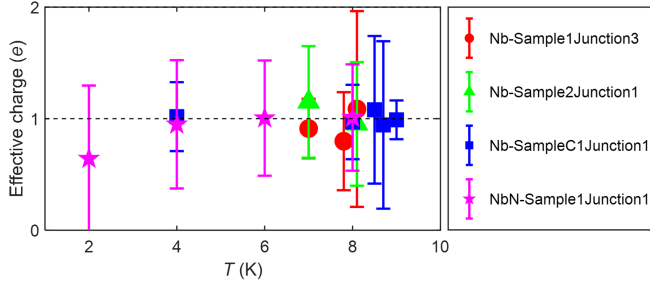


FIG. 4. Summary of the averaged effective charge inside the superconducting gap for different samples as a function of the temperature. The zero resistance  $T_c$  is  $\sim 8$  K for Nb-based samples and 15.5 K for NbN-based samples. The effective charge of the Nb/Al – AlO<sub>x</sub>/Nb sample at 4 and 7 K is measured at 0.1 T.

temperatures, different magnetic fields, and different samples. Figure 4 clearly shows that the effective charge inside the gap is  $1e$ , even though all samples are superconducting. Details of samples and Fig. 4 are shown in Supplemental Material [21].

Our results raise the question why no  $2e$  charge transfer is observed in devices where the materials are clearly in their superconducting state. In theory [18,20,28], the appearance of Cooper pairs and Andreev reflections should yield doubled shot noise when the temperature is below  $T_c$  in superconducting tunnel junctions, as previously observed in STMs [13], nanowires [14], and break junctions [29].

One possibility to obtain  $1e$  noise: tunneling through bound states in the insulator [30,31]. Such an indirect tunneling leads to the possibility of normal tunneling inside the gap. However, it also leads to signatures in the tunneling spectra. As we do not observe these, we believe that this possibility is unlikely to be solely responsible for our results. Instead, we argue here that this apparent inconsistency is due to the junction properties of typical mesoscopic setups, which are very different from those of STM [12,13,23] or nanowire [14]. In these systems, there is usually only one tunneling channel, and it is easy to deduce the transparency  $\tau$  from  $\tau = (G_0 R_j)^{-1}$  where  $G_0$  is the conductance quantum and  $R_j$  is the sample resistance. In contrast, our samples are mesoscopic tunnel junctions made by cleanroom fabrication methods and have larger junction areas, e.g.,  $25 \mu\text{m}^2$  for our SIS device. Because of this large junction area, the number  $N$  of tunneling channels in our sample is large, and it is *a priori* not possible to separately deduce the number of channels and the transparency  $\tau = (N G_0 R_j)^{-1}$ .

It is important to point out that mesoscopic junctions generally have large numbers of channels  $N$  with much smaller transparencies compared to pointlike junctions with the same resistance. And indeed, such a situation can lead to vastly different shot noise, as we show by simulations (Fig. 5). The key point is that the shot noise is controlled by the transparency  $\tau$ . While a typical single-channel junction with typical resistances will show  $2e$  noise, a mesoscopic

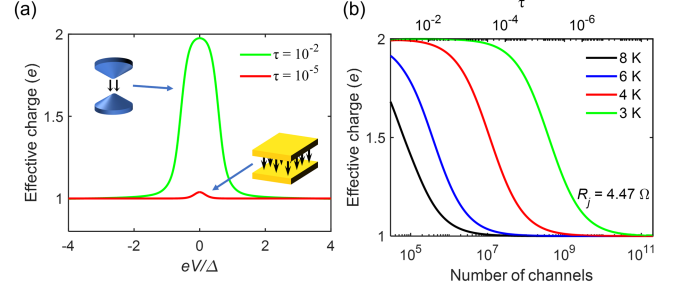


FIG. 5. Obscuring of  $2e$  noise in mesoscopic device geometries. (a) Effective charge calculated using BTK theory for transparencies of  $10^{-2}$  (typical for STM geometries; green line) and  $10^{-5}$  (typical for mesoscopic devices; red line). (b) Effective charge inside the gap as a function of the number of channels (bottom axis) and transparency (top axis) for a sample with  $4.47 \Omega$  normal state resistance for different temperatures.

junction with typical parameters (but without pinholes) will show  $1e$  noise, because of the vastly different transparencies. This can be illustrated with an example: A  $30 \Omega$  mesoscopic junction with an area of  $0.82 \mu\text{m}^2$  might have  $N \sim 10^7$  channels, meaning that the transparency is roughly  $10^{-5}$ , if homogenous channel parameters are assumed. It is these small transparencies that explain our  $1e$  noise. At finite temperatures, Andreev reflections and a normal tunneling process will happen at the same time inside the gap, and the transparency will control the contribution from each of these two processes to the total noise. The contribution from the normal process is proportional to  $\tau$ , while the contribution from Andreev reflections is proportional to  $\tau^2$ . Thus, when the transparency is low, the normal process will dominate the shot-noise signal, giving  $q = 1e$ ; only when the transparency is high, one can observe the noise enhancement. We generalize this further in Fig. 5(b) where we show simulations of the effective charge as a function of  $N$  or  $\tau$  for different temperatures. We use the parameters corresponding to our SIN sample. This simulation confirms that for an increasing number of channels (decreasing transparency), the effective charge decreases and provides guidelines for obtaining  $2e$  shot noise, as outlined below.

To determine where our experimental system is within this parameter range and test our hypothesis, we estimate the possible number of channels and their transparencies in our samples. The number of channels can be estimated by  $N = A * (k_F^2/\pi)$ , where  $A$  is the area of the junction and  $k_F$  is the Fermi wave vector [32].  $k_F$  can be estimated from  $k_F = (3\pi^2 n)^{1/3}$ , where  $n$  is the charge number density in the bulk. The charge densities of Nb [33] and NbN [27] are estimated from the literature. In Table I, we present our estimates for the numbers of channels and transparencies of our samples. When comparing these values with our model [Fig. 5(b)], we find that indeed our samples should yield  $q = 1e$  values.

TABLE I. Transparency estimation.

Sample	Nb/AlO <sub>x</sub> /Nb		NbN/oxide/Ag
	Samples 1, 2	Sample C1	Sample N1
Charge density	$5.56 \times 10^{28} \text{ m}^{-3}$		$1.60 \times 10^{29} \text{ m}^{-3}$
$R$	$31.6 \Omega$	$63.1 \Omega$	$4.47 \Omega$
Area	$0.82 \mu\text{m}^2$	$25 \mu\text{m}^2$	$0.09 \text{mm}^2$
$\tau$	$1.12 \times 10^{-5}$	$1.85 \times 10^{-7}$	$3.70 \times 10^{-10}$
$N$	$3.64 \times 10^7$	$1.11 \times 10^9$	$7.80 \times 10^{12}$

Thus far, we used BTK formulas to calculate the effective charge in SIN junctions. For SIS junctions, the situation is similar. We use a quasiparticle tunneling model [34] to calculate the contribution from  $q = 1e$  channels and find that  $q = 1e$  channels already dominate the tunneling processes due to the small  $\tau$  in our sample (see Supplemental Material [21]). We also note that there exist multiple Andreev reflections in SIS junctions, which will lead to further enhanced shot noise. However, multiple Andreev reflections processes will be suppressed compared to Andreev reflections at low transparencies, as their probabilities scale with  $\tau^n$ .

We can now use our model to estimate  $\tau$  and compare its prediction to different results reported in the literature. We find that  $q > 1e$  was reported in all systems where we expect large  $\tau$  ( $\tau > \sim 10^{-4}$ ) [12–15,35], and  $q = 1e$  was reported in most systems with small  $\tau$  [15,36], in agreement with our model (for details, see Fig. S9 in the Supplemental Material [21]). The exception stems from an experiment on a cuprate junction, where  $q > 1e$  was reported for a very small value of  $\tau$  [17]. It is an open question how to interpret this Letter; here we note that theory for noise of preformed pairs is still developing, and that the charge transfer layers can yield extra noise [37]. We also performed experiments on a YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> high- $T_c$  superconductor junction [38]; however, the noise spectrum in these samples is dominated by  $1/f$  noise (for details, see Fig. S8 in the Supplemental Material [21]).

In summary, we measured shot noise in SIS (Nb/Al – AlO<sub>x</sub>/Nb) and SIN (NbN/oxide/Ag) superconducting tunnel junctions in a high-frequency bandwidth (1.1–1.5 MHz) using a new, custom-built, high-resolution noise measurement system. We found that the measured effective charge equals  $1e$  both outside and inside the energy gap at temperatures below  $T_c$  for both types of junctions. This could be considered counterintuitive, as in principle, the noise in the superconducting state should correspond to  $q = 2e$  because of Andreev reflections. We interpreted our findings by proposing the presence of a large number of very small transparency channels in our sample, which obscure the pairing effect in noise measurements. BTK simulations quantitatively agreed with this picture. We further argued that this is in fact a common situation for mesoscopic junctions.

Our findings therefore indicated that to measure electron pairs without superconductivity in mesoscopic junctions— as opposed to single-channel STM junctions—one needs to carefully engineer a junction with few channels with very high transparency. Junctions with very thin insulator layers and clean interfaces are good candidates to achieve such a scenario. Further, pinholes in junctions, which are usually to be avoided, may give extra high transparency ( $q = 2e$ ) channels.

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