

Transparency-controlled multiple charge transfer in superconducting junctions with local shot-noise scanning tunneling spectroscopy

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Charge transport in superconducting junctions at finite voltages is governed by Andreev reflections, including multiple Andreev reflections, which are processes that enable multiple charge transfer, a hallmark that shot noise can directly quantify. Since the effective charge extracted from shot-noise measurements varies with the transparency of the junction, systematic control of transparency is essential but experimentally challenging. Here, we present shot-noise scanning tunneling microscopy measurements enabled by a newly developed amplifier, allowing access to different transparency regimes. We perform shot-noise measurements on Pb(111) with tunable transparency at 2.2 K and observe that the shot noise evolves from a single-electron tunneling regime to multiple charge-transfer regimes as transparency increases. Our results are quantitatively consistent with theoretical simulations of Andreev reflections and multiple Andreev reflections for a single-channel system. These results establish junction transparency as the key parameter governing the evolution of charge transport and demonstrate that noise STM is a powerful platform for investigating microscopic charge transport mechanisms with controlled junction transparency at the atomic scale.

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I. INTRODUCTION

In superconducting junctions, charge can be transferred in units larger than a single electron via multiple-charge transport processes. For example, in normal metal–insulator–superconductor (NIS) junctions Andreev reflection (AR) occurs when a bias voltage V lower than the superconducting gap Δ is applied across the junction. In this process, an electron in the normal electrode cannot enter the superconducting electrode as a single quasiparticle; instead, it is converted into a Cooper pair inside the superconducting region while a hole is reflected, resulting in an effective transfer of charge $2e$ [Fig. 1(a)]. In superconductor–insulator–superconductor (SIS) junctions, the tunneling current can be mediated by

multiple Andreev reflections (MARs), a process effectively transferring multiple charges across the junction. A MAR process of order n corresponds to an effective charge transfer of ne [1] and occurs at low tunneling junction transparency (τ) in the voltage range $2\Delta/n < e|V| < 2\Delta/(n-1)$ as shown in Fig. 1(b).

The effective charge transferred across a junction can be directly probed by shot-noise measurements [2]. Shot noise, which arises from the discrete nature of charge carriers, manifests as fluctuations in the electrical current. In the case of single-electron tunneling, the noise is Poissonian, with a zero-temperature power spectral density $S_I = 2qI$ in the low transparency limit, where $q = e$ is the carriers' effective charge and I is the average current. In systems with electron correlations or coherent transport [3–10], however, the effective charge of carriers q can deviate from $1e$. Hence, shot noise provides direct information about the effective charge of the tunneling current. It has long been employed in mesoscopic devices, where they enabled the detection of fractional charges in fractional quantum Hall systems [11,12] and the observation of ARs in NIS junctions [7,13], as well as MARs in SIS mesoscopic junctions [3–6]. More recently, noise spectroscopy combined with scanning tunneling

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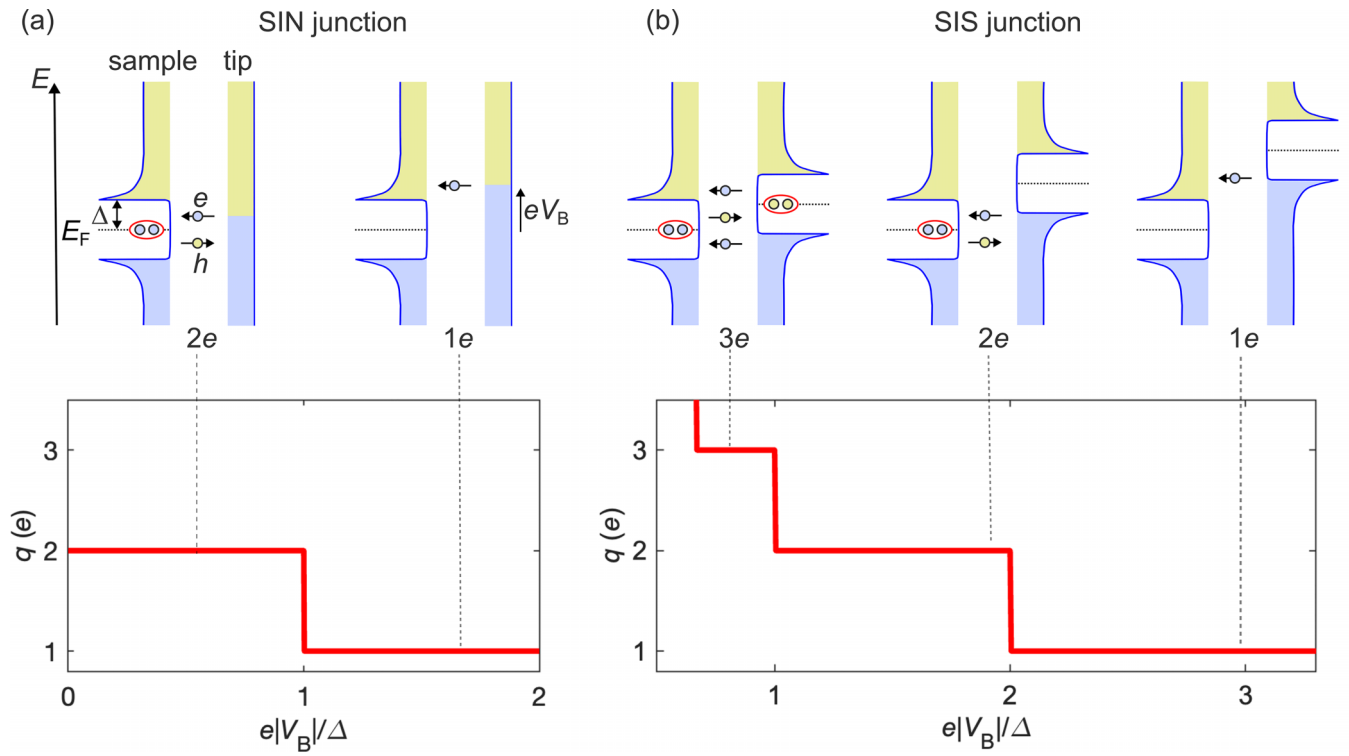


FIG. 1. Schematics of charge transfer in (a) SIN and (b) SIS junctions. The upper panels show sketches of the density of states of two electrodes, while the lower panels show the effective transferred charge q as a function of applied bias voltage V_B . (a) In an SIN junction, the normal current at $|eV_B| > \Delta$ is carried by quasiparticles transferring a single-electron charge. For $|eV_B| < \Delta$, an Andreev reflection process occurs: An electron transfers a Cooper pair into the superconducting condensate by reflecting a hole in the opposite direction, effectively transferring the $2e$ charge. (b) In an SIS junction, the tunneling current is dominated by multiple ARs (MARs) of order n , leading to an effective charge transfer of ne in the voltage range $2\Delta/n < |eV_B| < 2\Delta/(n-1)$.

microscopy (noise STM) has extended these measurements to the atomic-resolution, local tunneling junctions, allowing the detection of AR with spatial resolution [8–10].

The transparency of the tunneling junction plays a crucial role in determining the contribution of the different charge-transfer processes to the transport. AR and MAR processes rely on repeated transmission across the junction and are therefore highly sensitive to the junction transparency [14]. In the low-transparency regime ($\tau \lesssim 0.01$), the probability of an n th-order Andreev process scales as τ^n . Under these conditions, single-electron tunneling can become the dominant process, especially in the presence of thermal or quasiparticle lifetime broadening [15,16]. Such broadening can introduce quasiparticles within the superconducting gap (quasiparticle poisoning), making the $1e$ process more favorable and reducing the measured effective charge to $q \approx e$ even within the superconducting gap [16,17], where an ideal (M)AR dominated transport would yield $q > e$. Moreover, in the case of typical planar junctions, the large number N_{ch} of low-transparency tunneling channels averages out higher-order processes, effectively washing out any effect beyond single-electron tunneling [18]. On the other hand, in the higher transparency regime ($0.01 \lesssim \tau \ll 1$), the MAR process deviates from $q = ne$ expected in the lower τ regime; q becomes larger than ne in the energy range of $2\Delta/n < e|V| < 2\Delta/(n-1)$ as τ increases because the contributions of higher-order (more than n) MAR processes dominate [19–21].

Despite several experimental studies of MAR-related shot noise [3–6], a broadly tunable and well-controlled single-channel platform has been lacking. While quantitative agreement has been demonstrated in well-defined cases, such as aluminum atomic point contacts [4], the lack of a well-controlled system with tunable transparency has limited systematic shot-noise investigations in the single-channel limit. This motivates our experiments.

Scanning tunneling microscopy (STM) is ideal for this purpose because it provides a well-controlled tunneling junction. The junction transparency can be tuned continuously by changing the tip-sample distance. By combining shot-noise spectroscopy, STM enables access to detailed information about charge dynamics in the tunneling current across an atomically defined junction [22–24]. This technique has been successfully employed to detect the presence of electron pairs in both s -wave and d -wave superconductors [8,9,17,25]. When using STM, we can avoid the complexities associated with multiple transport channels that are typically present in planar junctions. In addition, the clean vacuum barrier intrinsic to STM junctions is free from the chemical and structural disorder of solid-state tunneling barriers. We can also precisely control the junction transparency. This transparency (τ) can be adjusted by tuning the tip-sample distance (d). τ can be estimated by $(G_0 R_{J,N})^{-1}$, where G_0 is the conductance quantum $2e^2/h$, h is Planck's constant, and $R_{J,N}$ is the normal state junction resistance. In STM measurements, the

tip-sample distance is stabilized via the setpoint current $I_{T,S}$ and set-bias voltage V_s that is normally chosen from values outside superconducting gap, which gives us $R_{J,N} = V_s/I_{T,S}$. A smaller $R_{J,N}$ is obtained by reducing d , which increases τ .

Here, we apply noise STM to superconducting Pb(111) at a temperature of 2.2 K and control the transparencies. First, in the SIN configuration, we observe that the effective charge inside the superconducting gap gradually increases as R_J decreases, corresponding to an increase in transparency τ . This indicates that at low τ , thermally excited quasiparticles contribute significantly to the tunneling process even within the gap. Next, we measure the shot noise of an SIS junction and detect effective charges larger than $2e$. We observe quantization of the effective charge and an R_J dependence consistent with the MAR process.

II. METHODS

We performed the measurements by using a customized low-temperature scanning tunneling microscope (USM-1500, Unisoku Co., Ltd). The surface of the Pb(111) is cleaned by repetitive cycles of Ar^+ sputtering at 1 kV with an Ar pressure of 5.0×10^{-5} mbar and subsequent annealing.

Our noise measurement apparatus has been described in detail in previous work [22]. Our custom-built cryogenic megahertz amplifier system consists of an RLC tank circuit and a high-electron-mobility transistor (HEMT) that converts the current fluctuations in the junction into voltage fluctuations across a 50Ω line, a room-temperature commercial amplifier (FEMTO HAS-X-1-40), and a spectrum analyzer (Zurich MFLI). For each bias voltage V_B applied to the sample, we will obtain the tunneling current $I_T = V_B/R_J$, where R_J is the STM junction resistance, and the shot-noise power $S_I = 2qI_T \coth(qV_B/2k_B T)$, where q is the effective charge and k_B is the Boltzmann constant. S_I will be first translated to a voltage noise S_V by the LC resonance circuit and then amplified by low-temperature and room-temperature amplifiers. Thus, the noise measured by a spectrum analyzer, S_V^m , is

$$S_V^m = G_{\text{total}}^2 |Z_{\text{tot}}^2| (S_I + S_I^{\text{off}1}) + S_V^{\text{off}0}, \quad (1)$$

where G_{total} is the total gain of the amplifier chain; $S_V^{\text{off}0}$ is the effective voltage noise background of the amplifier chain and the spectrum analyzer; $S_I^{\text{off}1}$ is the effective current noise background of the low-temperature amplifier and the resonator circuit, including the input current noise of the low-temperature amplifier, the Johnson noise of the tank circuit, and other additional noise sources effectively in parallel with the tank circuit; and Z_{tot} is the total impedance of the LC circuit and STM junction. We use the double tank circuit model representing the actual circuit because we use two LC tanks in parallel as the input impedance of the low-temperature amplifier. The total impedance is described as

$$Z_{\text{tot}} = ((Z_{\text{tank}} + Z_{\text{couple}})^{-1} + Z_{\text{tank}}^{-1} + R_{J,\text{diff}}^{-1})^{-1}, \quad (2)$$

where $Z_L = i\omega L$ is the impedance of one inductor, $Z_r = r$ is the resistance of the coil, and $Z_C = 1/i\omega C$ is the impedance of the total effective parallel capacitance $Z_{\text{couple}} = 1/i\omega C_{\text{couple}}$ ($C_{\text{couple}} = 100$ pF), and $R_{J,\text{diff}} = (dI/dV)^{-1}$ is the STM junction differential resistance. For more details about this double

tank circuit, see our recent work [22,23]. We record the experimental noise spectral density $S_V^m(f)$ across the full 100 kHz acquisition window (32 768 points) and fit the resonance curve's entire line shape to the functional form of Eq. (1), with L , C , and r to extract $S_I + S_I^{\text{off}1}$. The full frequency dependence of $|Z_{\text{tot}}(f)|^2$ is determined by the fit itself, and shot noise and thermal noise from the STM junction (S_I) and background noise ($S_I^{\text{off}1}$) entering only as a single frequency-independent amplitude. In the fitting procedure, we used a fixed known value for $L = 66 \mu\text{H}$, and C and r as fitting parameters. All capacitive elements are lumped in parallel with the LC tanks. Therefore, the fitted effective capacitance C includes not only the tank capacitance but also any other contributions like the cable capacitance that are not explicitly represented as a separate circuit element. We also obtain the resistance r by the fitting because the value cannot be directly measured. r is nearly constant across the data range in this paper. For more details, see Sec. I of the Supplemental Material [26].

Next, the effective charge can be calculated by

$$2qI_T \coth\left(\frac{qV_B}{2k_B T}\right) = S_I = (S_V^m - S_V^{\text{off}0}) / (G_{\text{total}}^2 |Z_{\text{tot}}^2|) - S_I^{\text{off}1}. \quad (3)$$

G_{total} and $S_I^{\text{off}1}$ ($\sim 1690 \text{ fA}^2/\text{Hz}$) are calibrated on a Au(111) surface and calibrated again by using data at bias larger than the gap on Pb(111) to avoid tiny change due to the fluctuation in the environment. In addition, Z_{tot} is calibrated for data at each bias using the shape of the resonance peak in the frequency domain. The shot-noise power at each V_B is averaged for at least 9 min with 1500 spectra.

The error bars in S_I are defined as the sum of two different errors in our measurements. The first is determined in the same way as in our previous works [16]—the fluctuation of the current noise in time, measured before measurements, yields a standard deviation of $1.72 \text{ fA}^2/\text{Hz}$. This contribution originates from our technical measurements setup, and therefore it is independent of any setting parameters such as V and I_T . The second arises from the effect of low $R_{J,\text{diff}}$ on the resonance curve fitting. In most cases, $R_{J,\text{diff}} \gg |Z_{\text{tank}}|$ and the circuit response is dominated by the tank circuit (the Q factor $\sim 1/|Z_{\text{tot}}|$), essentially independent of $R_{J,\text{diff}}$. However, when $R_{J,\text{diff}} < |Z_{\text{tank}}|$, $Q \sim R_{J,\text{diff}}$, indicating the resonance curve becomes broader and the mismatch between our model and the real circuit becomes more pronounced, increasing the uncertainty of the fit. To solve this problem, we introduce a special effective gain calibration process to compensate for this mismatch although the real HEMT gain is constant. However, the effect is negligible for most of our data, and we retain only the data for which the uncertainty remains small. We only show the data with large uncertainty that need this special calibration process in Fig. S4 of the Supplemental Material [26]. For further details about this calibration and uncertainty, see Sec. I in the Supplemental Material [26]. The error bars in q correspond to numerical solutions of Eq. (3), using the upper and lower bounds of the error bars in S_I . Since $q \simeq S_I/2I_T = S_I R_J/2V$, even if the error bar of S_I is constant, the error bar of q becomes larger with a decrease in I_T or V .

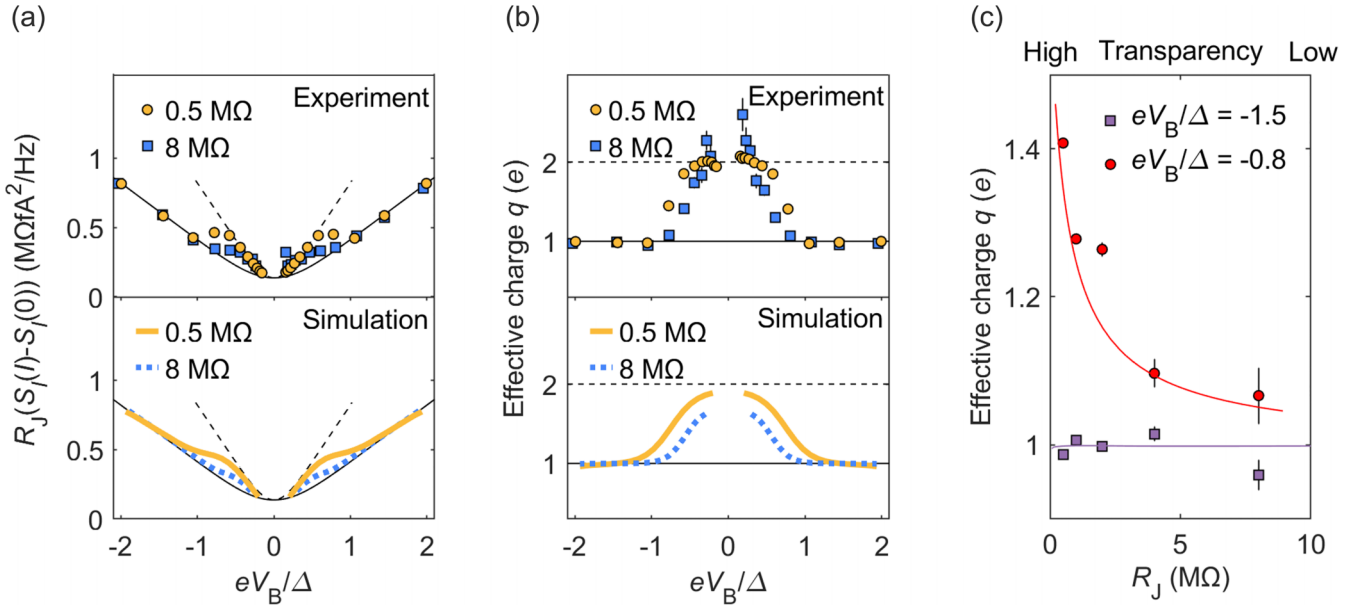


FIG. 2. R_J -dependent noise measurements in SIN junction. (a) Measured (top) and simulated (bottom) current noise taken with two typical R_J . See Sec. III in the Supplemental Material for more results [26]. The solid (dashed) line represents the calculated values for effective charge equals $1e$ ($2e$). (b) Effective transferred charge q extracted from (a). Note that at small bias, the effective charge can be larger than $2e$, which could be related to the unstable tip at small bias. (c) Effective transferred charge q at $eV_B/\Delta = -1.5$ and -0.8 as a function of R_J . The solid lines correspond to simulated values.

III. RESULTS

A. SIN junction

First, we use a PtIr tip and Pb sample to form a SIN junction. Figure 2(a) shows the shot noise S_J as a function of bias voltage V_B taken with two typical $R_J = V_B/I_T$. It is clear that shot noise shows transparency-dependent behavior. For more results with different R_J , see Sec. III of the Supplemental Material [26]. Note that each S_J curve is obtained at fixed R_J with the STM feedback loop active during the V_B sweep (constant- R_J mode). This data acquisition mode is different from the usual method for acquiring dI/dV curves in STM, where the feedback loop is turned off after stabilizing the tip height (constant-height mode) and then sweeping V_B to obtain dI/dV signals. For details, see Sec. I and Fig. S6 in the Supplemental Material [26]. In constant-height mode, the transparency τ is constant with V_B sweep. In contrast, in constant- R_J mode, the tip height can change with V_B , and therefore, τ is also V_B dependent. The advantages of the constant- R_J mode are that it is less sensitive to mechanical noise and allows one to detect the small signals inside the superconducting gap.

From the data shown in Fig. 2(a), we can extract the effective charge $q = S_J/2e$ as shown in Fig. 2(b). As expected, $q = 1e$ outside the superconducting gap, i.e., for $|eV_B| > \Delta$, where $\Delta = 1.28 \text{ meV}$ obtained by fitting the dI/dV spectrum with the Dynes formula [27] (see Sec. I in the Supplemental Material [26]). The noise signal starts to increase at a bias voltage of $|eV_B| \sim \Delta$ and saturates around $2e$ for voltages deep inside the gap. The transition between $1e$ and $2e$ becomes sharper for lower R_J (larger τ). As shown in Figs. 2(a) and 2(b), the experimentally observed R_J dependence can be explained by our simulations using the Blonder-Tinkham-Klapwijk (BTK)

framework [15,28,29]—a single-channel theoretical model. The BTK model is based on the Bogoliubov–de Gennes equations and describes the tunneling current for a NS junction with an arbitrary barrier strength (related to τ), in the framework of scattering matrix formalism. More details on the simulation are given in Sec. II of the Supplemental Material [26]. Figure 2(c) shows how the value of the effective charge inside and outside the gap ($eV_B/\Delta = -0.75$ and -1.5 , respectively) depends on R_J . The BTK simulations accurately reproduce the experimentally observed behavior. The value of the effective charge does not reach $q = 2e$ for $eV_B/\Delta = -0.75$ due to single-particle ($1e$) contributions to the tunnel current, enabled by the finite temperature of the experiment, as well as lifetime broadening, in combination with the low transparency. These results demonstrate the importance of the tunneling transparency τ .

B. SIS junction

Next, we measure the noise S_J in a SIS junction and extract the effective charge q . Figure 3(a) shows our constant-height dI/dV results on a Pb(111) surface with a Pb tip. The superconducting tip is prepared by indenting the PtIr tip into the clean Pb(111) surface [30–32]. We fit the results and obtained superconducting gap amplitude $\Delta = 1.32 \text{ meV}$, where we assume the gap of the tip and the sample are the same, and quasiparticle lifetime broadening $\Gamma = 0.04 \text{ meV}$. Figures 3(b) and 3(c) show S_J and constant- R_J dI/dV measured at $R_J = 500 \text{ k}\Omega$. Note that the constant- R_J dI/dV is obtained together with the noise, which is different from constant-height dI/dV [Fig. 3(a)] that is often obtained in STM measurements. For details of constant- R_J dI/dV , see Fig. S7. We observe peaks at $eV_B = 2\Delta/n$ in the constant- R_J dI/dV curve and

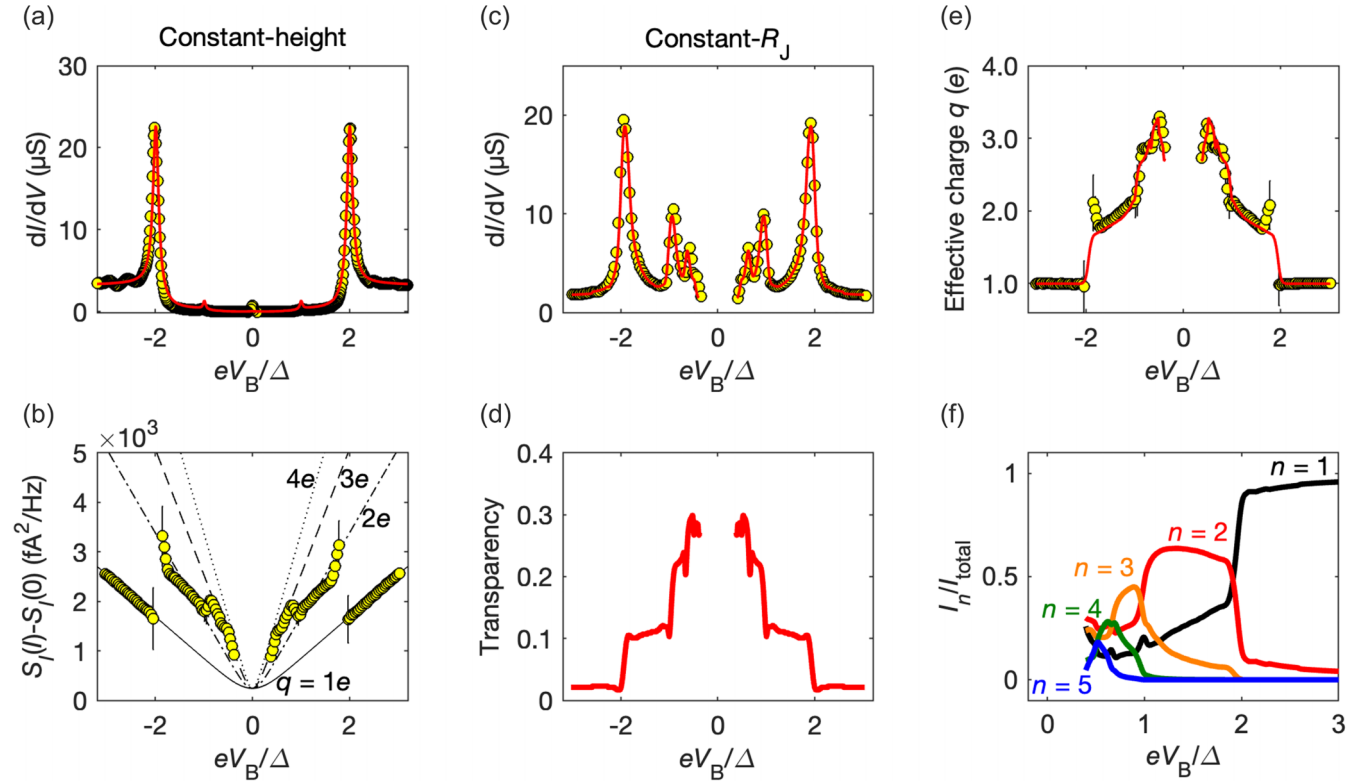


FIG. 3. Noise measurements in SIS junction. (a) Constant-height differential conductance (dI/dV) spectrum. The tip height is stabilized with the tunneling condition of $V_S = 5$ mV and $I_T = 10$ nA. The red curve shows a fitted function with $\Delta = 1.32$ meV and $\Gamma = 0.04$ meV. (b) Measured current noise with constant $R_J = 500$ k Ω . The curves are the expected noise from $2qI$ with an effective charge q of $1e$, $2e$, $3e$, and $4e$. The error bar is determined by the fluctuation of the current noise in time before each experiment, yielding a standard deviation, and by the uncertainty in the calibration of the cryogenic amplifier associated with the resonator circuit including the STM junction. We retain only the data for which the uncertainty remains small. See Sec. I and Fig. S3 for further details and Fig. S4 for the full data set in the Supplemental Material [26]. (c) Measured dI/dV with constant $R_J = 500$ k Ω simultaneously obtained with (b). (d) By fitting the constant- R_J dI/dV [the red curve in (c)], we can extract transparency as a function of V_B . (e) Effective charge q obtained from (b). The red curve is calculated by using $\Gamma = 0.04$ meV and effective transparency (e). (f) The contribution of n th-order MAR with $q = ne$ obtained from the analysis. We use a measurement temperature of $T = 2.2$ K for all calculations.

corresponding jumps in the S_I curve. The additional peaks in the constant- R_J dI/dV spectra are indicative of MAR processes taking place. As shown in Fig. 3(e), the extracted $q(V_B)$ shows a multistep behavior, and reaches values $q > 3e$. This is consistent with the occurrence of MAR.

The $q(V_B)$ curve does not show the clean steplike structure presented in Fig. 1(b). This deviation is due to MAR processes of different order occurring simultaneously. First, we have $q < ne$ in $\Delta/n < e|V_B| < 2\Delta/n$, which is obvious, especially for $n = 4$, where q decreases as V_B decreases. The reduction can be due to the contribution of excited quasiparticles, which allows lower-order MAR processes ($n < 4$). Second, we have noise enhancement with decreasing V_B , accompanied by an effective charge q exceeding $2e$ in the range $\Delta < e|V_B| < 2\Delta$. The reason for this is that transparency is large enough for higher-order multiple Andreev reflection processes to contribute significantly. To understand this quantitatively, we extract the bias-dependent transparency by fitting the experimental constant- R_J dI/dV [the red curve in Fig. 3(c)] using the single-channel MAR theory [33], where all orders of MAR processes are naturally

included. The extracted transparency is shown in Fig. 3(d) and it was then used to compute the noise with the help of the theory [19]. The result for the bias-dependent charge obtained by using the extracted transparency, Δ and Γ , is shown as a red curve in Fig. 3(e), which is in agreement with the effective charge extracted from the measurements. To obtain information about the dominant transport processes, we utilize the concept of full counting statistics [20,21] and compute the charge-resolved current contributions due to the different tunneling processes for the parameters extracted from our fits. We show the results in Fig. 3(f). Note that, as expected, MARs dominate the transport for subgap voltages, but the single-quasiparticle tunneling (I_1) still has a finite contribution even at voltages below 2Δ because of the quasiparticle broadening and the finite temperature. In addition, we note that the results around $e|V_B| \sim 2\Delta$ are not shown in Figs. 3(b) and 3(e), since $R_{J,\text{diff}} = (dI/dV)^{-1}$ is too low in this regime; the resulting resonance noise spectral density from our RLC tank circuit shows a resonance that is too broad, and the uncertainty of the fit used to extract S_I is large, as discussed in the Methods section. See Fig. S4 in the Supplemental Material for the full data set [26].

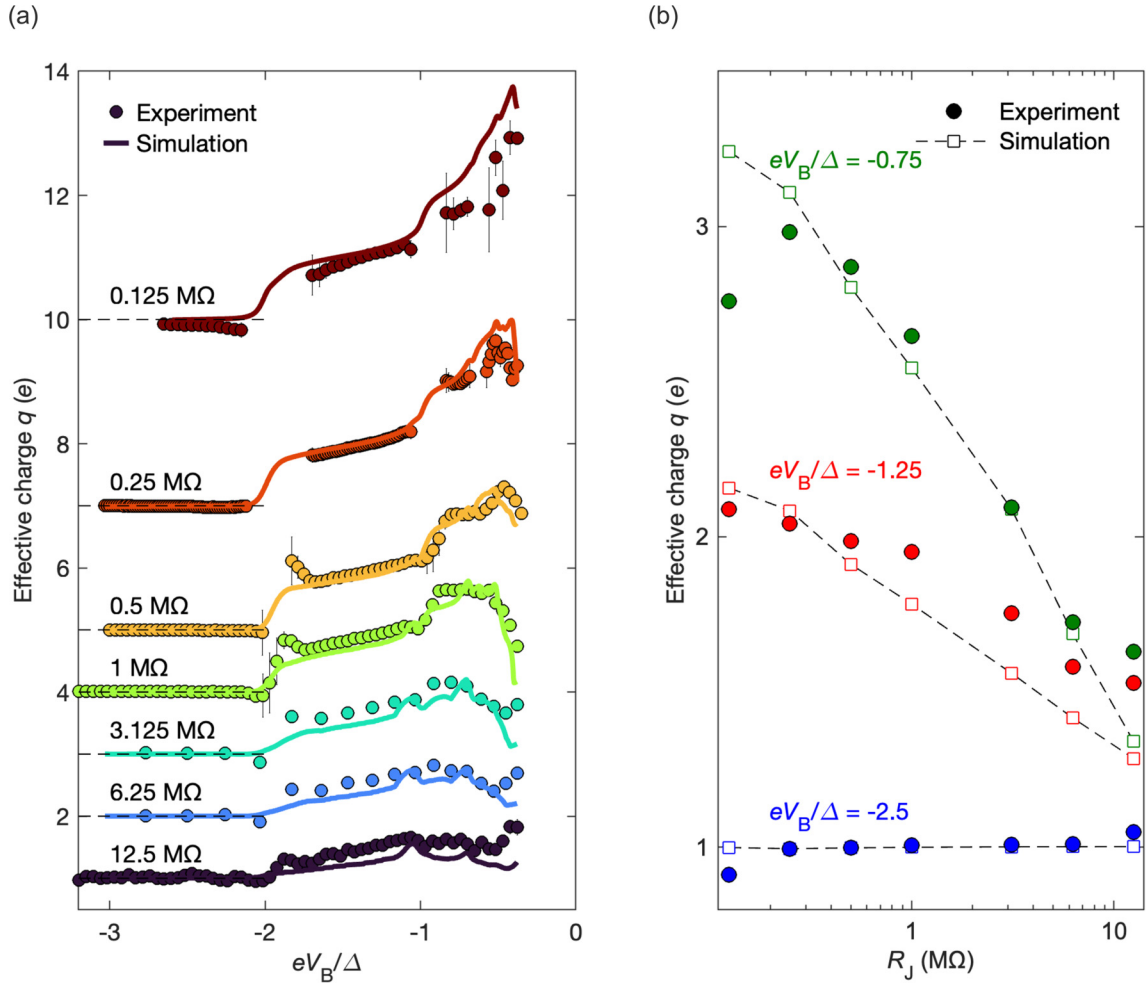


FIG. 4. R_J -dependent noise measurements in an SIS junction. (a) Measured (circles) and simulated (solid lines) effective charge q taken with different R_J . The results are shifted by a vertical offset for clarity and dashed lines indicate $q = 1e$ with the offset. The error bars are determined in the same way as in Fig. 3(b). We retain only the data for which the uncertainty remains small. See Sec. I and Fig. S3 for further details and Fig. S4 for the full data set in the Supplemental Material [26]. (b) Dots correspond to the effective charge q at $eV_B/\Delta = -2.5$ (blue), -1.25 (red), and -0.75 (yellow) as a function of R_J . The squares denote simulated values and dashed lines serve as guides to the eye.

To further confirm that the observed noise originates from MAR processes, we perform noise measurements with different R_J as shown in Fig. 4(a). In each region of $2\Delta/n < eV_B < 2\Delta/(n-1)$, q becomes larger with lower R_J , which is also reproduced by our simulation (solid lines). Figure 4(b) shows measured q (filled circles) and simulated (open squares) values of q for different voltages as a function of R_J . For higher R_J , q is smaller than ne , whereas for lower R_J , q is enhanced and exceeds ne . We conclude that lower R_J suppresses contributions from excited quasiparticles that allows lower-order MAR processes and enhances contributions from higher-order MAR processes. We note that our R_J control measurements did not exhibit the expected noise reduction below $1e$ associated with the Fano factor $1 - \tau$ at $eV_B > \Delta$ for the SIN junction and $eV_B > 2\Delta$ for the SIS junction. This is because in these bias-voltage regimes, the noise enhancement arising from (M)AR also increases with τ . For example, Fig. 3(f) shows that I_2 remains finite even at voltages above 2Δ . Thus, the expected noise reduction is very small and falls below our experimental resolution, unless a bias voltage $eV_B \gg \Delta$

is applied. In practice, however, experimentally we cannot reach much higher bias voltage with low enough junction (differential) resistance while maintaining stable tunneling conditions.

IV. CONCLUSION

In summary, we have demonstrated shot-noise measurements of SIN and SIS junctions with noise STM in a tunneling regime, enabling a direct comparison with single-channel theoretical models of Andreev processes. We clearly observe $q = 2e$ in SIN and $q = ne$ in SIS junctions from the multiple charge transport. Our data are in quantitative agreement with single-channel simulations and confirm transparency as a key parameter governing the evolution of charge transport: increasing τ enhances higher-order MAR processes, leading to q exceeding ne , while decreasing τ suppresses q toward $1e$. This highlights the importance of transparency for engineering and probing correlated charge transport.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [34].

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