## **Coherent Multiple Charge Transfer in a Superconducting NbN Tunnel Junction**

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We measured shot noise and submillimeter-wave response in a superconducting NbN tunnel junction that had a subharmonic gap structure on the current-voltage (*I-V*) curve. We found that the observed effective charge, defined from the noise-current ratio, tends to a steplike function of voltage. In the presence of submillimeter-wave radiation of frequency  $\nu$ , novel step structures spaced by  $h\nu/2e$  below and above the half-gap voltage clearly appeared on the *I-V* curve, overlapping the ordinary photonassisted tunneling steps spaced by  $h\nu/e$ . Observation of these features provides clear evidence that coherent multiple Andreev reflection processes occur in the NbN tunnel junction with low barrier transparency.

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Since the subharmonic gap structure (SGS) in superconductor-insulator-superconductor (SIS) tunnel junctions was first observed by Taylor and Burstein in 1963, the phenomenon's physical mechanism has been open to discussion [1]. The origin of SGSs is now thought to be multiple Andreev reflections (MAR) when the barrier transparency is higher than the tunnel limit [2]. Recently, the microscopic theory of MAR [3] has predicted its role is even more important: MAR dominate electronic transport in mesoscopic systems. In a MAR process of order m(where *m* is an integer), multiple charges of *me* are transferred at threshold voltages of  $2\Delta/me$ . Under a dc bias, the shot noise can reveal information on the transport of large charge quanta,  $q(\gg e)$ , at subgap voltages [4]. This nature has been observed in a tunnel junction [5] and atomic point contacts [6]. To extract the intrinsic shot noise from the total observed noise, complicated calculations must be made with measured dc parameters. Thus, this is a rather indirect method of observing MAR processes.

Another method of investigating MAR processes in a junction is to observe the frequency response in the presence of electromagnetic radiation. Under the timedependent potential, the photon energy,  $h\nu$ , can assist the transport of a charge through a multiphoton process. For an ideal tunnel junction with charge transfer e, current steps appear at voltages of  $(2\Delta + nh\nu)/e$  (where n is an integer) on the current-voltage (I-V) curve because of the photonassisted tunneling (PAT) effect [7]. If multiple charges (me) transfer due to MAR, subharmonic PAT steps at voltages of  $(2\Delta + nh\nu)/me$  should be observed, and the theoretical analysis recently predicted that the step structure will be pronounced at lower transparencies [8]. The appearance of these steps spaced by hn/me in the voltage directly provides information on MAR processes as a visible quantum effect on the I-V curve. Despite this interesting feature, investigations of MAR processes in superconducting junctions under electromagnetic irradiation are rare.

NbN tunnel junctions have been developed [9] for use as sensitive heterodyne mixers in the submillimeter-wave band [10]. The junctions have SGSs that have been well characterized [11] by the semiclassical theory of Ref. [2]. However, the charge transfer process in junctions is essential to a better understanding of MAR. In this Letter, we present investigations of MAR processes in an NbN tunnel junction under a dc bias and rf excitation at submillimeterwave frequencies. The observations of shot noise and rf-induced steps suggest that coherent multiple charge transfer due to MAR occurs in the NbN tunnel junction with low transparency. We believe that we have made the first clear observation of subharmonic PAT steps resulting from photon-assisted MARs in a tunnel junction, which is a clearer and more direct evidence of MAR processes than observing shot noise alone.

A typical high-quality NbN/MgO/NbN junction with a critical current density of 6.8 kA/cm<sup>2</sup> was used in our experiment. It was mainly fabricated using reactive dcmagnetron sputtering and partly using rf sputtering in a load-locked sputtering system [12]. The junction size was 10.8  $\mu$ m long by 0.6  $\mu$ m wide so that it behaved as a distributed element at submillimeter-wave frequencies for a broadband response [13]. The junction and a 2-section impedance transformer connected to it were integrated into a lens-coupled twin-slot antenna to efficiently couple the incoming electromagnetic waves to the junction [14]. The dc and rf characteristics of the junction were investigated using the heterodyne receiver system described in Ref. [15]. A backward wave oscillator was used as a local oscillator (LO) to introduce a monochromatic rf signal, and a blackbody at room temperature or cooled in liquid nitrogen was used as the other rf signal. In the receiver system, the junction is connected via an isolator to 1.25-1.75 GHz low noise amplifiers and bandpass filters to observe the intermediate frequency (IF) output of the rf signals at a given dc bias. Without the rf signals, the IF output gives information about the shot noise generated in the junction.

With the rf signals, the IF output was used instead of making the usual dV/dI measurement to determine the step structures in the voltage induced on the *I-V* curve because the IF output had information about the dynamic resistance. A magnetic field was applied to the junction by using a superconducting Nb coil to suppress the Josephson current and the associated rf-induced steps. All measurements were performed at a receiver bath temperature of 4.2 K.

First, we made detailed measurements of the shot noise generated in the NbN tunnel junction without LO irradiation. The IF output of the receiver and the current through the junction as a function of bias voltage are shown in Fig. 1(a). The superconducting gap is clearly visible at a bias of  $2\Delta$ (= 5.6 meV) because of the ordinary single quasiparticle current rise on the *I*-*V* curve and the dip in the IF output. The dips indicated by arrows in the subgap IF output locate the threshold voltages of eV =  $2\Delta/m$  (*m* = 2, 3) for the corresponding SGS in the current caused by MAR. The IF output of the receiver can be calibrated in



FIG. 1. (a) Measured and calculated IF output of the NbN SIS receiver as a function of voltage without LO irradiation. The IF output was calibrated to equivalent noise temperature based on Eq. (1). Effective charge *e* was assumed for the calculated one. The SGS is clearly visible up to the third harmonic in the measured IF output. Also shown are the measured current through the NbN junction and the assumed leakage current as a function of voltage. A resistance of 500  $\Omega$  in parallel with the junction was assumed for the leakage current, which was not related to MAR. (b) The calculated effective charge of the NbN junction as a function of voltage. Assuming the leakage current, the dependence of  $1 + 2\Delta/eV$  was derived.

terms of the equivalent Rayleigh-Jeans temperature at the IF amplifier input by comparing with the theoretical prediction [16]

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$$T_{\rm eq} = \frac{1}{4k_B} S_I R_{\rm dyn} (1 - \Gamma^2) + T_{\rm iso} \Gamma^2 + T_{\rm IF}, \qquad (1)$$

where

$$S_I = 2qI \tag{2}$$

and

$$\Gamma^2 = \left| \frac{R_{\rm dyn} - R_{\rm iso}}{R_{\rm dyn} + R_{\rm iso}} \right|^2.$$
(3)

The term  $R_{dyn}$  is the differential resistance (dV/dI) of the junction at a dc bias,  $R_{iso}$  is the 100  $\Omega$  impedance of the matching load for the isolator using a 180° hybrid coupler [17],  $\Gamma$  is the reflection coefficient between  $R_d$  and  $R_{iso}$ ,  $T_{\rm iso}$  is the physical temperature of the isolator (=4.2 K),  $T_{\rm IF}$  is the equivalent noise temperature of the IF chain, and  $S_I$  is the shot noise spectral density. In the theoretical calculation using Eq. (1), we assumed single quasiparticle tunneling [q = e in Eq. (2)]. The calibration was made based on the facts that  $q \simeq e$  at voltages above the gap voltage and that  $\Gamma^2 \simeq 1$  at the gap voltage [18], which determined that  $T_{\rm IF} = 5.5$  K. The  $R_{\rm dyn}$  we used in our calculation was obtained by numerical differentiation of the experimental *I-V* data shown in Fig. 1(a). Clearly, below the gap voltage, the measured noise is larger than the calculated one. Next, we calculated the voltage dependence of the effective charge q(V) in the NbN tunnel junction by dividing the measured spectral density of the junction shot noise by 2eI. A theoretical tunneling current below the gap voltage due to thermal excitation can be ignored in the measured current,  $I_{\text{meas}}$ , because the operating temperature of 4.2 K is much lower than the junction's critical temperature of about 15.7 K. The calculated results are shown in Fig. 1(b). Although the effective charge was almost constant at twice the electron charge below the gap voltage when  $I_{\text{meas}}$  was directly used in the calculation, the  $1 + (2\Delta/eV)$  dependence due to MAR [4] was obtained by assuming a leakage current caused by a parallel resistance,  $R_{\text{leak}}$ , of 500  $\Omega$  (which has a negligibly small equivalent thermal noise temperature). In fact, when this is done, a steplike  $(1 + Int[2\Delta/eV])$  dependence of the effective charge on bias voltage is seen [19], which was more pronounced than a previously reported result for another NbN tunnel junction [5]. This evidence suggests that the tunnel barrier transparency is in the low transmission regime. Although the occurrence of MAR processes in the junction was found by the shot noise investigation under a dc bias, it was necessary to extract a "true" current related to MAR from the measured current with adjustable parameters. This is an unavoidable uncertainty in dc measurements for tunnel junction devices.

To directly observe multiple charge transfer due to MAR in the NbN tunnel junction, we measured the frequency

(4)

response in the presence of submillimeter-wave radiation. The rf excitation should interact with the current carried by MAR processes with the aid of the PAT effect. Because of the quantum interaction of multiple photon energies of  $nh\nu$ with multiple charge transfer of me due to coherent MAR processes in the junction, a subharmonic PAT step appears on the *I*-V curve at a voltage of  $V = (2\Delta + nh\nu)/me$  [8]. The I-V characteristics of the NbN tunnel junction, obtained with and without LO irradiation of 625 GHz in the receiver are shown in Fig. 2(a), and the IF output power as a function of bias voltage for hot and cold loads with LO is shown in Fig. 2(b). The I-V curve with LO shows the usual PAT steps resulting from the charge transfer of single quasiparticles. The steps in the dc voltage have a spacing of  $h\nu/e$  (= 2.57 mV), both below (absorption of photons) and above (emission of photons) the gap voltage of  $2\Delta/e$ . Overlapping these ordinary steps, subharmonic PAT steps with a spacing of  $h\nu/2e$  were clearly observed above and below the half-gap voltage of  $2\Delta/2e$ , and these corresponded to the dips in the IF output for hot and cold loads. This evidence directly suggests that an effective charge of 2e was transferred at a threshold voltage of  $2\Delta/2e$  because of coherent MAR processes at low transmission, as described by Cuevas et al. [8]. Therefore, we may use higher order perturbation theory to expect how this structure evolves with the LO irradiation power. Since the junction



FIG. 2. Characteristics of the measured current and IF output as a function of voltage with LO irradiation of 625 GHz. (a) *I*-V curve of the NbN junction. Subharmonic PAT step structures spaced by  $h\nu/2e$  in voltage can be seen around  $2\Delta/2e$ . (b) IF output of the receiver. Locations of dips in the curve determine the voltage of current step structures induced by LO irradiation.

was long (about twice the guided wavelength,  $\lambda_g$ , of 625 GHz), the *m*th order current step height at a voltage of  $(2\Delta + nh\nu)/me$  is assumed to be [20]

 $I_{m,n}^{\text{step}} = L^{-1} \int_0^L J_n^2 [meV_{\text{LO}}(X)/h\nu] dX I_m,$ 

where

$$V_{\rm LO}(X) = V_{\rm LO0} |\cosh(\gamma X)|. \tag{5}$$

The term  $J_n(x)$  is an ordinary Bessel function of order n,  $I_m$ is the current step height responsible for the mth SGS without LO irradiation, L is the length of the junction,  $V_{I,O}(X)$  is a spatial variation in LO voltage caused by a standing wave [21], X is the distance along the junction from its open end,  $V_{\rm LO0}$  is the voltage amplitude at the open end of the junction, and  $\gamma$  is the propagation constant of the junction. For a small junction  $(L \ll \lambda_g)$ , the term m = 1corresponds to the result of the Tien-Gordon theory. To verify Eq. (4) experimentally, we investigated the relationship between the height of ordinary PAT step at  $(2\Delta$  $h\nu)/e$  (m = 1 and n = -1) and subharmonic PAT step at  $(2\Delta - h\nu)/2e$  (m = 2 and n = -1) by changing the LO irradiation power at 625 GHz. Since both steps were sharply defined as shown in Fig. 2, the height of each step was measured by taking the vertical distance at the step voltage between tangent lines to the *I-V* curve above and below the step [21], and was normalized to the measured current step height responsible for the *m*th order without LO power. The experimental relationship between the step heights is shown in Fig. 3. For the theoretical calculation, we assumed the spatial variation pattern of the LO voltage as shown in the inset in Fig. 3, which was



FIG. 3. Relationship between ordinary PAT current step heights at  $(2\Delta - h\nu)/e$  and subharmonic PAT current step heights at  $(2\Delta - h\nu)/2e$  for various LO irradiation powers at 625 GHz. Each current height is normalized to current height at threshold voltage without LO irradiation. Inset shows the assumed standing-wave pattern in the junction. The theoretical relationship was calculated when the excitation strength of  $eV_{LOO}/h\nu$  was varied from 0 to 2.



FIG. 4. Voltage positions of subharmonic PAT steps as a function of LO frequency. Solid lines show the predicted voltage positions of subharmonic steps:  $V = (2\Delta \pm h\nu)/2e$ .

derived from the experimental junction parameters. Although the theoretical prediction still included some assumptions, the LO power dependence of the subharmonic PAT step height was distinctly different from that of the ordinary PAT step height under the same variation of rf excitation as described by the perturbation theory (where the height tends to  $J_n^2(m\alpha)$ ,  $\alpha$ : excitation strength). This behavior may be another hallmark of a photon-assisted MAR process in the tunnel junction with low transparency.

The most convincing evidence of photon-assisted MAR features is the dependence of the voltage position of the step on the frequency of LO irradiation. If our observed subharmonic PAT steps around the half-gap voltage at 625 GHz were caused by coherent MAR processes in the junction, we should observe the steps at voltages of the theoretical prediction according to  $V = (2\Delta \pm h\nu)/2e$  at any given frequency. To investigate the frequency dependence of the steps, we measured the voltage positions of the steps induced on the *I-V* curve by monitoring the corresponding dips in the IF output curve at various frequencies of LO irradiation from 600 to 800 GHz. We plotted the voltage positions of the subharmonic PAT steps against the irradiated frequency as shown in Fig. 4. The frequencies were precisely determined by  $\nu = \delta V e/h$ , where  $\delta V$  is the measured spacing in voltage for the ordinary PAT steps below the gap voltage. For the measured frequency range, the data points fall neatly on the theoretical prediction of  $V = (2\Delta \pm h\nu)/2e$  with no adjustable parameters. This frequency-dependent feature clearly shows that the observed step structures under the rf excitation result from photon-assisted MAR in the NbN tunnel junction with low transparency. Accordingly, our observation of the subharmonic PAT steps provides direct evidence that multiple charges are transferred in the junction, which is a further signature of the coherent MAR processes under the rf excitation.

In summary, we have measured shot noise enhancement caused by the steplike behavior of the effective charge as a function of voltage in a superconducting NbN tunnel junction under a dc bias, and this behavior is attributed to a coherent MAR process in the junction with low transparency. As direct evidence of this process, we have observed subharmonic PAT effects in the presence of submillimeterwave radiation. Both the frequency- and pump-strength dependences of the PAT steps associated with an effective charge of 2e display the hallmarks of photon-assisted MAR processes at low transmission. To our knowledge, this is the first clear observation of coherent MAR processes under a time-dependent potential in a superconducting tunnel junction.

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