Coherent Low-Energy Charge Transport in a Diffusive S-N-S Junction

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(March 22, 2022)

We have studied the current voltage characteristics of diffusive mesoscopic Nb-Cu-Nb Josephson junctions with highly-transparent Nb-Cu interfaces. We consider the low-voltage and high-temperature regime $eV < \epsilon_c < k_B T$ where $\epsilon_c$ is the Thouless energy. The observed excess current as well as the observed sub-harmonic Shapiro steps under microwave irradiation suggest the occurrence of low-energy coherent Multiple Andreev Reflection (MAR).

PACS : 73.23.-b, 74.50.+r, 74.80.Fp

A S-N-S junction made of a normal metal (N) embedded between two superconducting electrodes (S) exhibits a zero-voltage Josephson supercurrent [1]. At the N-S interface, the microscopic mechanism is the Andreev reflection of an electron into a hole which traces back almost exactly the trajectory of the incident electron. This coherent process corresponds to the transfer of a Cooper pair in the normal metal. Let us consider a mesoscopic diffusive normal metal where the length $L$ is larger than the elastic mean free path $l_e$ but smaller than the phase-coherence length $L_\varphi$. In the long junction regime $L \gg \sqrt{D/\Delta}$ of interest here, the relevant energy scale is the Thouless energy $\epsilon_c = h/\tau_D = hD/L^2 \ll \Delta$. Here $D = v_F l_e/3$ is the diffusion coefficient, $\tau_D = L^2/D$ is the diffusion time, $\Delta$ is the superconducting gap. The essential fact is that Andreev pairs with a low energy obeying $\epsilon < \epsilon_c$ remain coherent over the whole normal metal length $L$. This coherent window contributes significantly to phase-coherent transport even if the thermal distribution width $k_B T$ is much larger than the Thouless energy $\epsilon_c$. This is exemplified in Andreev interferometers by the large magnetoresistance oscillations with an amplitude of about $\epsilon_c/k_B T$. Such a weak power-law temperature dependence is in striking contrast with the exponential decay of the Josephson coupling.

At finite bias voltage ($V$), a dynamic regime takes place where the superconducting phase is time-dependent while successive Andreev-reflected electrons and holes gain the energy $eV$ at each travel. This Multiple Andreev reflections (MAR) process leads to a sub-gap structure at the voltages $2\Delta/pe$ (p integer) in the current-voltage characteristic [2] and the energy distribution function [3]. In quantum point contacts, the whole process of MAR is phase-coherent since the electron transit time is very short. This regime of coherent MAR was recently emphasized in the description of both the current voltage characteristics, the shot noise and the d.c. supercurrent [4] of ballistic atomic contacts [5]. The related non-sinusoidal current-phase relation is indicative of coherent multiple charge transfer, but no direct observation has been reported so far.

In a diffusive S-N-S junction, the diffusion of electrons through the normal metal involves the diffusion time $\tau_D$ which must be compared to the period of the phase difference $\chi$ evolution $\tau_V = h/2eV$. The quasi-static condition for a small phase evolution during $n$ successive Andreev reflections at each interface writes $2n\tau_D < \tau_V$, which is equivalent to $eV < \pi \epsilon_c/2n$. In this regime, electrons and holes in the coherent window ($\epsilon < \epsilon_c$) can experience MAR while maintaining global coherence of the induced Andreev pairs. This process should imply the coherent transfer of multiple charges as in short ballistic point contacts, provided the interface transparency is close to 1. Recent experiments in diffusive S-N-S junctions focused on the opposite regime $\epsilon_c < eV \approx \Delta$ [4]. Subgap structures were observed, but no signature of coherent MAR was found.

The time evolution of the out-of-equilibrium energy distribution induced by MAR was also considered phenomenologically, taking into account the energy relaxation. This lead to the prediction of an enhancement of the junction conductance at finite voltage [1-2]. The related a.c. Josephson coupling at twice the Josephson frequency was experimentally confirmed [4]. A close analogy can be drawn with the dissipative transport in a mesoscopic Aharonov-Bohm ring [1-4]. Indeed both the relaxation current in such a ring and the MAR excess current in a S-N-S junction appear at bias voltages below $h/\tau_e$ [1-3] where $\tau_e$ is the energy relaxation time.

In this Letter, we present an investigation of the coherent dynamic regime in high-quality diffusive S-N-S junctions. We focus on the contribution of low-energy Andreev pairs to the current-voltage characteristic. We used as a probe an a.c. microwave field with a small photon energy $h\omega < \epsilon_c$ so that it sits within the coherent energy window. Our main result is the observation of sub-harmonic Shapiro steps that we discuss in terms of coherent multiple pair transfer at low energy. Here, the relevant number of Andreev reflections $h\omega/2eV$ is imposed by the photon microwave energy, not by any energy gap.

The samples consist of a small Cu conductor attached to two Nb electrodes which are superconducting below
$T_c \approx 8$ K (Fig. 2a inset). The electrical contacts include an on-chip capacitance (0.2 pF) which connects the microwave circuit made of a cryogenic 50 $\Omega$ coaxial cable to the sample. The fabrication process makes use of e-beam lithography and a shadow deposition technique based on a thermostable resist compatible with UHV electron-beam Nb evaporation [13]. The two samples described here (labelled A and B) belong to the series of S-N-S junctions investigated in Ref. [16]. They show a similar behavior. We unintentionally kept the thermal fluctuations negligible at every temperature by designing wide junctions so that their normal-state conductance $G_N$ and hence the Josephson energy $E_J = \hbar I_c/2e$ is large: $E_J > k_BT_c$. The sample and measurement parameters were also chosen to fulfill the condition $\epsilon_c < k_BT < \Delta$. The parameters for sample A (respectively B) are the following: length of the normal metal $L = 710 (800)$ nm, width 580 nm and thickness 100 nm. The diffusion coefficient is $D = 250 (230)$ cm$^2$/s and the normal state resistance is 0.152 (0.183) $\Omega$. The calculated Thouless energy $\epsilon_T = 33 (24)$ $\mu$eV coincides with the values obtained from the magnitude and temperature dependence of the d.c. critical current $I_c$ assuming perfect transmission at the S-N interfaces [10].

Fig. 1a shows the differential resistance of sample A as a function of the d.c. current bias. The critical current $I_c$ is easily identified by the sharp jump in differential resistance, even at high temperatures when it is strongly reduced. The differential resistance characteristics show a striking behavior above 4 K: the amplitude of the main peak at the critical current decreases while a broader bump develops at higher current. In this temperature range the current-voltage is fully reversible, which discards heating effects. The comparison in Fig. 1a with the ohmic behaviour and the resistively shunted junction (RSJ) model $V = G_N^{-1} \sqrt{T^2 - I_c^2}$ (at 5.5 K) demonstrates that this behavior corresponds to a low-energy excess current. The observed features are reminiscent of the foot-like structure previously observed in superconducting micro-bridges [17,18] which was as reminiscent of the foot-like structure previously observed in superconducting micro-bridges [17,18] which was as-

We investigated the coherent nature of the low voltage electrical transport by applying a low power (-20 dBm) microwave field. This low power ensures that the microwave acts only as a probe and does not drive the electron energy distribution. Fig. 2a shows the differential resistance and the current as a function of voltage for sample B at 4 K in the presence of an a.c. current $I_{\omega}$ of frequency 6 GHz. The feature corresponding to the Shapiro step is observed at voltage $V_1 = \hbar \omega/2e = 12.4$ $\mu$V (index 1 on the figure), as expected. In addition, we observe two structures at one half: $V_{1/2} = 6.2$ $\mu$V and one third: $V_{1/3} = 4.1$ $\mu$V of this voltage (index $\frac{1}{2}$ and $\frac{1}{3}$). The latter is clearly visible only on the differential resistance curve. Because of the high temperature and the small a.c. excitation, the plateau amplitudes are small and rounded by thermal fluctuations. The magnitude of these structures was observed to increase linearly with the microwave current $I_{\omega}$, leading to true plateaux with
zero differential resistance at high a.c. power (-10 dBm). Then, a large number of peak structures, both harmonic and sub-harmonic, emerge from the noise level. We found that their location is independent on both the temperature and the microwave power. The position of the observed peak structures as a function of the microwave frequency between 4 and 18 GHz is plotted in Fig. 3. The harmonic 1 represents the fundamental Shapiro step. The ensemble of the peaks obeys the simple law given by 

\[ V = m/n \cdot \hbar \omega /2e \]

While the integer steps \( (n = 1) \) are simple results of the ordinary Josephson coupling, the observation of fractional steps \( (n > 1) \) deserves a detailed discussion.

In order to extract the step amplitudes, we assume that the steps can be described by the RSJ model: the step width is twice an effective critical current \( i_{m/n} \), smoothed by a thermal broadening. We used either an analytical expression or the high temperature approximation to determine for each temperature and each step the actual half width \( i_{m/n} \). Fig. 4 summarizes its temperature dependence for the three main Shapiro steps \((1/2, 1/2, 1)\) at 6 GHz microwave frequency. At the center of the step 1 plateau, the current bias \( I \simeq G_N V_1 \) is much larger than the critical current \( I_c \), so that the step amplitude should follow the usual voltage-bias law \( i_1 = I_c J_1(I_1/I) \). Here \( J_1 \) is the Bessel function of the first kind. As can be seen in Fig. 4 the temperature dependence of the amplitude \( i_1 \) strictly follows that of \( I_c \). The small observed ratio \( i_1/I_c \approx 0.06 \) allows an estimate of the a.c. excitation current \( I_x \approx 10 \mu A \) at -20 dBm. The treatment of the microwave current as a non-perturbative probe is hence fully justified. The most striking observation is the non-monotonic temperature dependence of both \( i_{1/2} \) and \( i_{1/2} \), in strong contrast with the monotonous behavior of \( i_1 \).

As seen in Fig. 4, \( i_{1/2} \) starts increasing at temperature of about 3 K, then reaches the same amplitude as \( i_1 \) near 4 K, and finally slowly decreases at higher temperatures. Let us point out that the temperature at the maximum is much larger than the Thouless temperature \( \epsilon_c/k_B = 0.28 \). A similar behavior is found on the smaller \( 1/3 \) step. The persistence of the \( 1/3 \) and \( 1/2 \) at high temperatures, i.e. when the d.c. Josephson coupling is suppressed, is the most important result of this work.

![FIG. 2. Lower curve (right scale): Differential resistance of sample B at 4 K as a function of voltage in presence of a microwave current of frequency 6 GHz and power -20 dBm. Structures induced by the microwave current appear at the usual Shapiro step position \( V_1 = \hbar \omega /2e = 12.4 \) \( \mu V \) and also at one half and one third of this value. Upper curve: the corresponding voltage steps on the current-voltage characteristic.](image1.png)

![FIG. 3. Peak position as a function of frequency for integer (black symbols) and non-integer steps (open symbols) in sample B. These steps were observed at a 4K temperature and a -10 dBm microwave power.](image2.png)
scale because of the smaller electron transit time.

Let us now discuss the implications of sub-harmonic steps in terms of coherent MAR processes. The bias voltage involved here is so low that the phase difference is quasi-static and low-energy Andreev electron pairs are phase-coherent over the junction length. This is the regime where electrons close to the Fermi level experience coherent MAR. The $\frac{1}{2}$ step is therefore associated with the coherent transfer across the diffusive metal of double pairs. Along the same way, the $\frac{1}{3}$ step corresponds to the transfer of triple pairs. In the case of an interface transparency close to 1, this interpretation holds provided the phase-breaking events can be neglected. In this respect, the characteristic time scale $2n\tau_D = 2n \times 28$ ps has to be smaller or at least comparable to the phase coherence time $\tau_\phi \simeq 160$ ps at 4 K [20]. At this temperature, the condition is fulfilled for $n = 2$ and 3, which is consistent with our observations of $\frac{1}{2}$ and $\frac{1}{3}$ steps at low power. Higher-order steps up to $n = 5$ arise only at high power when the inelastic damping is compensated by a larger a.c. current.

In summary we have investigated the low-voltage coherent transport in diffusive S-N-S junctions with high barrier transparency. We observed a series of sub-harmonic Shapiro steps at high temperature and low voltage $eV < \epsilon_c < k_BT$ where ordinary Josephson coupling is known to vanish. Presently there is no satisfactory description of the microscopic mechanism which is very likely to involve coherent transfer of multiple charges.

We acknowledge fruitful discussions with Y. Blanter, M. Büttiker, J. C. Cuevas, K. Lehnert, A. Levy-Yeyati, F. Wilhelm, A. Zaikin and within the TMR program “Dynamics of superconducting nanocircuits”. We thank F. Hekking for pointing out the relevance of coherent MAR.

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