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

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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 ϵ_c is the Thouless energy. The observed excess current as well as the observed subharmonic Shapiro steps under microwave irradiation suggest the occurrence of low-energy coherent multiple Andreev reflection.

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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 S and its inverse to the diffusion of an electron Andreev 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_φ. In the long junction regime $L \gg \sqrt{\hbar D/\Delta}$ of interest here, the relevant energy scale is the Thouless energy $\epsilon_c = \hbar/\tau_D = \hbar D/L^2 \ll \Delta$. Here $D = v_F l_e/3$ is the diffusion coefficient, $\tau_D = L^2/D$ is the diffusion time, and Δ 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 [2]. 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 ϵ_c. This is exemplified in Andreev interferometers by the large magnetoresistance oscillations with an amplitude of about $\epsilon_c/k_B T$ [2]. 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 subgap structure at the voltages $2\Delta/pe$ (p integer) in the current-voltage characteristic [3] and the energy distribution function [4]. 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 current-voltage characteristics, the shot noise and the dc supercurrent [5] of ballistic atomic contacts [6–8]. The related nonsinusoidal 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 τ_D which must be compared to the period of the phase difference χ evolution $\tau_V = h/2eV$. The quasistatic 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 $\epsilon_c < eV \simeq \Delta$ [9,10]. 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 led to the prediction of an enhancement of the junction conductance at finite voltage [11,12]. The related ac Josephson coupling at twice the Josephson frequency was experimentally confirmed [13]. A close analogy can be drawn with the dissipative transport in a mesoscopic Aharonov-Bohm ring [14]. 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 $\hbar/\tau_e$ [11,12,14], where τ_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 ac microwave field with a small photon energy $\hbar\omega < \epsilon_c$ so that it sits within the coherent energy window. Our main result is the observation of subharmonic Shapiro steps that we discuss in terms of coherent multiple pair transfer at low energy. Here, the relevant number of Andreev reflections $\hbar\omega/2eV$ 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

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$T_c \simeq 8$ K (Fig. 1a inset). The electrical contacts include an on-chip capacitance (0.2 pF) which connects the microwave circuit, made of a cryogenic 50 Ω coaxial cable, to the sample. The fabrication process makes use of e-beam lithography and a shadow evaporation technique based upon a thermostable resist compatible with UHV electron-beam Nb evaporation [15]. The two samples described here (labeled A and B) belong to the series of S-N-S junctions investigated in Ref. [16]. They show a similar behavior. We intentionally 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: $E_J > k_B T_c$. The sample and measurement parameters were also chosen to fulfill the condition $eV < \epsilon_c < k_B T < \Delta$. 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) Ω. The calculated Thouless energy $\epsilon_c = 33(24)$ μeV coincides with the values obtained from the magnitude and temperature dependence of the dc critical current I_c assuming perfect transmission at the S-N interfaces [16].

Figure 1a shows the differential resistance of sample A as a function of the dc 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 characteristic shows 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. 1b with the Ohmic behavior and the resistively shunted junction (RSJ) model $V = G_N^{-1}\sqrt{I^2 - I_c^2}$ (at 5.5 K) demonstrates that this behavior corresponds to a low-energy excess current. The observed features are reminiscent of the footlike structure previously observed in superconducting microbridges [17,18] which was assigned to nonequilibrium processes in the presence of a gap oscillation. In our long S-N-S junctions, the role of the gap is played by the minigap which sets the energy scale at ϵ_c. The microscopic origin of the excess current is the coherent MAR: each Andreev reflection at the N-S interfaces transfers a Cooper pair in and out, until an inelastic process takes place [19]. On average, each coherent electron transfers τ_e/τ_D times the elementary charge. From the nonequilibrium model of Ref. [12], a differential resistance bump is predicted at $eV = \hbar/\tau_e$. In the experiment, the position of the differential resistance maximum corresponds to $\tau_e = 90$ ps at $T = 5.5$ K. This value is consistent with the known electron-phonon relaxation time in Cu [20] and exceeds the diffusion time $\tau_D = 20$ ps at every temperature. It gives a phase-coherence length $L_\varphi \approx 1.5$ μm [21] which is about twice the normal metal length. The amplitude of the conductance enhancement ∂G was estimated by Zhou and Spivak [11] as $\partial G/G_N \simeq (\epsilon_c/k_B T)(\tau_e/\tau_D)$. This gives here $\partial G/G_N \simeq 0.3$ at 5.5 K, in qualitative agreement with the experiment.

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. Figure 2 shows the differential resistance and the current as a function of voltage for sample B at 4 K in the presence of an ac current I_ω of frequency 6 GHz. The feature corresponding to the Shapiro step is observed at voltage $V_1 = \hbar\omega/2e = 12.4$ μV (index 1 on the figure), as expected. In addition, we observe two structures at one-half, $V_{1/2} = 6.2$ μV and one-third, $V_{1/3} = 4.1$ μV 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 ac 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_ω, leading to true plateaus with zero differential resistance at high ac power (-10 dBm). Then, a large number of peak structures, both harmonic and subharmonic, emerge from the noise level. We found that their location is independent of 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

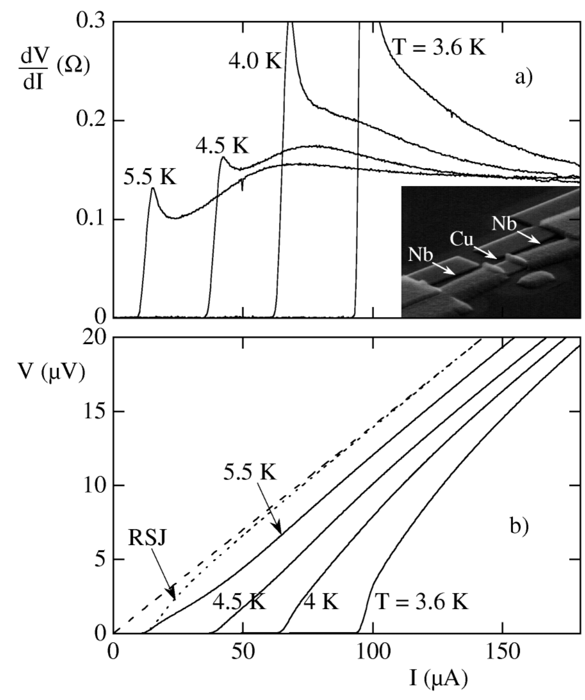

FIG. 1. (a) Measured differential resistance of sample A as a function of dc bias current at different temperatures. Inset: electron micrograph of a typical sample made by shadow evaporation. (b) Current-voltage characteristics as obtained by numerical integration of differential resistance curves. Also shown is the theoretical curve calculated from the RSJ model at 5.5 K (dotted line) and the Ohmic behavior (dashed line). The data all sit in the coherent window $eV < \epsilon_c = 33$ μeV.

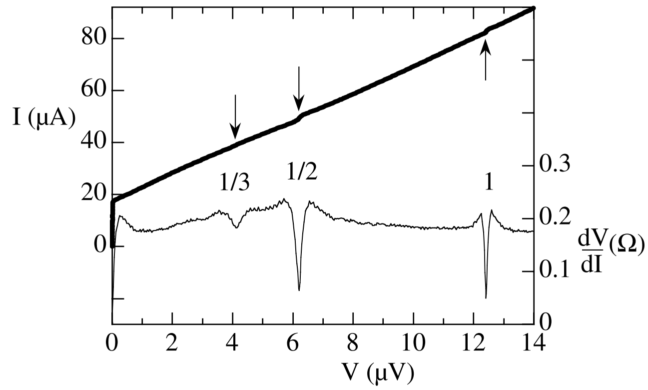

FIG. 2. Lower curve (right scale): differential resistance of sample B at 4 K as a function of voltage in the presence of a microwave current of frequency 6 GHz and power -20 dBm. Structures induced by the microwave current appear at the usual Shapiro step position $V_1 = \hbar\omega/2e = 12.4\ \mu$V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

in Fig. 3. The harmonic 1 represents the fundamental Shapiro step. The ensemble of the peaks obeys the simple law given by $V = (m/n)(\hbar\omega/2e)$. 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 the effective critical current $i_{m/n}$ smoothed by a thermal broadening. We used either an analytical expression or a high temperature approximation [1] to determine for each temperature and each step the actual half width $i_{m/n}$. Figure 4 summarizes its temperature dependence for the three main Shapiro steps ($1, \frac{1}{2}$, and $\frac{1}{3}$) 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 $i_1 = I_c J_1(I_\omega/I)$ [22]. 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 ac excitation current $I_\omega \approx 10\ \mu$A at -20 dBm. The treatment of the microwave current as a nonperturbative probe is hence fully justified. The most striking observation is the nonmonotonic temperature dependence of both $i_{1/2}$ and $i_{1/3}$, in strong contrast with the monotonous behavior of i_1. As seen in Fig. 4, $i_{1/2}$ starts increasing at a 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 $\epsilon_c/k_B = 0.28$ K. Similar behavior is found on the smaller $\frac{1}{3}$ step. The persistence of the $\frac{1}{2}$ and $\frac{1}{3}$ at high temperatures, i.e., when the dc Josephson coupling is suppressed, is the most important result of this work.

The persistence of (integer) Shapiro-like steps at high temperatures [23] is expected to occur in a four-terminal configuration under current injection from a normal reservoir, but not in our two-terminal geometry. One can also discard the inductance or the capacitance of the junction itself or the onset of chaos as a possible origin of subharmonic steps [1], as our high-conductance junctions are strongly overdamped. The subharmonic steps must indeed be viewed as a manifestation of a nonsinusoidal Josephson current-phase $I(\chi)$ relation. An intrinsic nonsinusoidal contribution to the dc supercurrent is expected at zero temperature but strongly suppressed as the temperature increases [24]. In a long junction at $k_B T > 5\epsilon_c$ (≈ 2 K for our samples) the second harmonic is expected to contribute to less than 2% of the total current. In contrast, the subharmonic steps observed here not only appear as the temperature increases, $T > 3$ K as seen in Fig. 4, but they eventually exceed the integer steps. The intrinsic

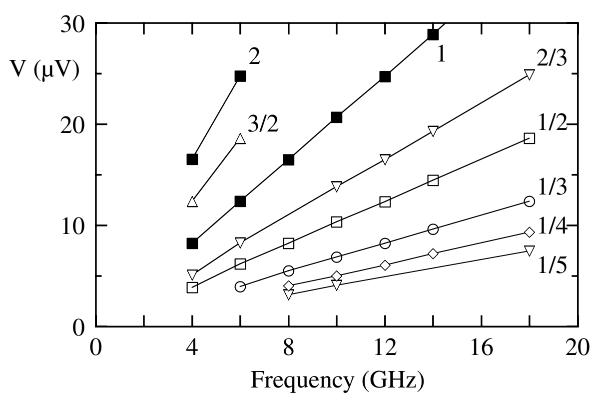

FIG. 3. Peak position as a function of frequency for integer (solid symbols) and noninteger steps (open symbols) in sample B. These steps were observed at a 4 K temperature and a -10 dBm microwave power.

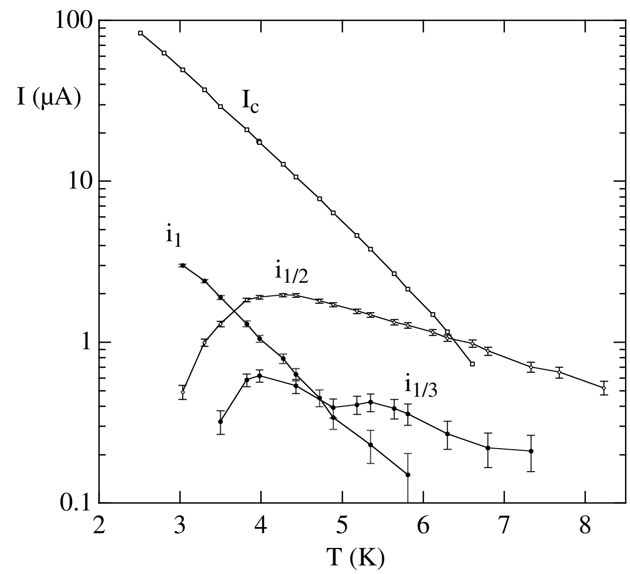

FIG. 4. Temperature dependence of the half width $i_{m/n}$ of integer and fractional steps in sample B under -20 dBm microwave irradiation at 6 GHz. The detection threshold was about 0.1 μA. The measured dc critical current I_c is plotted on the same graph.

nonsinusoidal phase dependence of the dc Josephson current therefore cannot explain the subharmonic steps. The nonequilibrium ac Josephson coupling model [12] introduced in our discussion of the excess current was shown to predict a finite $\sin 2\chi$ term even in the absence of dc Josephson critical current. Lehnert *et al.* [13] reported such an observation in InAs based junctions in the clean limit. Our results in the diffusive regime show strong similarities with [13] although the latter ones are concerned with higher electron energy scale because of the smaller electron transit time.

Let us now discuss the implications of subharmonic steps in terms of coherent MAR processes. The bias voltage involved here is so low that the phase difference is quasistatic 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 $\tau_\varphi \simeq 160$ 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 ac 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 subharmonic Shapiro steps at high temperature and low voltage $eV < \epsilon_c < k_B T$, 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.

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