Multiple Andreev Reflection and Giant Excess Noise in Diffusive Superconductor/Normal-Metal/Superconductor Junctions

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We have studied superconductor/normal metal/superconductor (SNS) junctions consisting of short Au or Cu wires between Nb or Al banks. The Nb based junctions display inherent electron heating effects induced by the high thermal resistance of the NS boundaries. The Al based junctions show in addition subharmonic gap structures in the differential conductance $dI/dV$ and a pronounced peak in the excess noise at very low voltages $V$. We suggest that the noise peak is caused by fluctuations of the supercurrent at the onset of Josephson coupling between the superconducting banks. At intermediate temperatures where the supercurrent is suppressed a noise contribution $\propto 1/V$ remains, which may be interpreted as shot noise originating from large multiple charges.

I. INTRODUCTION

Superconductor/normal-metal (SN) interfaces of high transparency exhibit remarkably different properties for electric charge and energy transfer, respectively. Quasiparticles with energy $\epsilon$ below the energy gap $\Delta$ of the superconductor cannot enter the superconductor. This implies a high thermal resistance of the SN boundary since energy is exclusively carried by the quasiparticles. In contrast, charge can be transmitted at $\epsilon < \Delta$ via the Andreev reflection process: An electron coming from the normal side is reflected as a hole and a Cooper pair is transferred to the superconductor. This should have important consequences for the energy distribution of quasiparticles in a short normal bridge connected to two superconducting reservoirs. Here we assume that the length $L$ of the bridge is larger than the thermal diffusion length $L_T = \sqrt{\hbar D/2\pi k_B T}$ which governs the penetration of Cooper pairs into the normal wire. For $L > L_T$ the supercurrent through the structure is exponentially weak.

In the case of normal reservoirs the distribution function $f(\epsilon)$ in the wire either assumes a two step shape if the inelastic scattering length $L_{in}(\epsilon) \gg L$, or smears out into a Fermi-Dirac function with a spatially varying electron temperature if $L_{in}(\epsilon) \ll L$. The broadening of $f(\epsilon)$ has been detected by local tunneling spectroscopy, or by measuring the power spectral density $S_V(V)$ of the current noise in the junction.

In the case of superconducting reservoirs the broadening of $f(\epsilon)$ is expected to be much more dramatic when compared to normal reservoirs. In particular for small applied voltages $eV \ll \Delta$ quasiparticles have to climb up to the energy gap $\Delta$ via multiple Andreev reflections (MAR) at the two SN boundaries in order to remove the deposited energy into the reservoirs. For samples shorter than the phase coherence length $L_\phi$ subharmonic gap structures have been observed in diffusive samples in the differential conductance $dI/dV$ at voltages close to $V = 2\Delta/en$ where $n$ counts the number of reflections in the MAR cycle. Such structures have been found earlier in superconducting microbridges, tunnel junctions and ballistic S/N/S point contacts. In addition, there are theoretical and experimental indications that the coherent MAR cycle transfers multiple charge quanta of magnitude $2\Delta/V$, which should lead to an enhanced current noise at low bias voltages. For diffusive systems so far only indications of charge doubling have been reported, which points towards single Andreev reflection events.

II. SAMPLE PREPARATION

The samples are prepared by angle evaporation through a suspended Si$_3$N$_4$ mask on a Si substrate with a SiO$_2$ spacer layer. The anorganic mask avoids the previously observed deterioration of the superconducting properties of the Nb by outgassing of the conventional organic resist (e.g. PMMA) during evaporation of the high melting point Nb. The Si$_3$N$_4$ top layer is patterned by conventional electron beam lithography. Wet etching of the 800 nm SiO$_2$ layer results in the desired undercut profile. The high mechanical strength of the Si$_3$N$_4$ allows large undercuts and freely suspended bridges of several micron length. The transition temperature of narrow Nb wires displayed only a minor reduction (0.2 K) of the superconducting transition temperature $T_c$ when compared to codeposited Nb films. Our SNS devices consist of thin ($\approx 15$ nm) normal wires (Au or Cu) of 0.4 - 2 $\mu$m length and 100 - 200 nm width between thick (50 - 200 nm) reservoirs made of Nb or Al. A scanning elec-
of the N metal. The spectral density $S$ of voltage fluctuations across the sample is measured as a function of current bias in the frequency range between 100 and 400 kHz with a cross-correlation technique. With this technique we obtain a voltage sensitivity of $50 \, \text{pV}/\sqrt{\text{Hz}}$ with commercial room temperature preamplifiers. The measurements are performed in a superconducting transition of the reservoirs. Further reduction of the temperature leads to a continuous decrease of $R$ which becomes more drastic below 2 K. This proximity induced reduction of $R$ is accompanied by a sharp peak in the differential conductance $dI/dV$ at zero bias voltage. The peak has a width of $50 \, \text{µV}$ (which is close to $k_B T$ at 0.3 K) and can be seen as the precursor of a supercurrent which emerges when $L_T$ becomes comparable to the wire length $L$. The peak has a height of only 10% of the normal state resistance $R_N$ of the wire for $L = 2 \, \text{µm}$, while we find supercurrents up to $50 \, \text{µA}$ for $L = 0.4 \, \text{µm}$ in samples with Nb banks.

In Fig. 3 we present the excess noise $S_V$ of a series of 9 Nb/Au/Nb junctions with 2 µm long Au wires. As a reference measurement, we first collected data in a perpendicular magnetic field of 6 T in which the Nb reservoirs are normal (open squares). For a direct comparison of the (effective) electron temperatures $T_{el}$ we have normalized $S_V$ with $dV/dI$ (see right-hand scale). For lower voltages $V < 1 \, \text{mV}$ the measured noise falls on the 1/3 reduced shot noise (dashed line). At higher voltages, additional cooling via electron-phonon scattering results in a negative curvature of $S_V$.

In the case of superconducting reservoirs (solid circles) we find a dramatic increase of $S_V$ in particular for the smallest voltages. The normalized excess noise rises with nearly vertical slope at $V = 0$ and merges at $V \sim 2\Delta/e$ into the 6 T curve. The latter is expected because for energies $\epsilon \gg \Delta$ the probability of Andreev reflection rapidly vanishes. Note that $T_{el}$ is already $\approx 6 \, \text{K}$ for $V \approx 2\Delta/e$. From weak localization measurements on the long Au wire we infer $L_\phi = 0.9 \, \text{µm}$ at 1.3 K (0.55 µm at 4.2 K). Since $L_\phi$ is considerably shorter than the wire length of 2 µm the MAR cycle is incoherent. This is confirmed by the absence of subharmonic gap features in $dI/dV$ (see Fig. 2).

The electron temperature in the Au wire is controlled by the power dissipation in the wire, the energy loss via
Electron-phonon (dashed line) and N/S interface (dotted line) electron heating effect according to Eqs. (1) and (2). Inset: contributions to the cooling power as a function of electron temperature $T_{el}$ in the wire. The solid line gives an estimate of the shot noise of noninteracting electrons in case of normal reservoirs. The solid line gives an estimate of the electron heating effect according to Eqs. (1) and (2). Inset: Electron-phonon (dashed line) and N/S interface (dotted line) contributions to the cooling power as a function of electron temperature $T_{el}$ in the wire. $T_{el}$ in the reservoirs is assumed to remain at 0.27 K.

quasiparticle transmission through the S/N interfaces, and the electron phonon scattering in the Au wire. At low $T_{el}$ the electronic heat diffusion within the Au wire is much faster than the energy loss across the interfaces so that we may assume local thermal equilibrium with a nearly constant temperature profile along the wire. The heat transfer through the interfaces can be reasonably well described in terms of a simple BTK-like expression for the heat current $P_{NS}(T_{el})$ through the N/S boundaries:

$$P_{NS}(T_{el}) = \frac{2}{R_{m} \epsilon^2} \int_{-\infty}^{\epsilon} \epsilon (f_{N} - f_{S}) (1 - A - B) d\epsilon. \quad (1)$$

Here, $R_{m}$ is the normal state resistance of the N/S boundary, $f_{N}(T_{el})$ and $f_{S}(T_{Bath})$ are the Fermi functions in the wire and the reservoirs, while $A(\epsilon, Z)$ and $B(\epsilon, Z)$ are the coefficients of Andreev- and normal reflection and $Z$ is the interface parameter. We estimate $R_{m} \approx 5 \Omega$. Within our simplified model, the cooling via electron phonon contribution scattering is given by

$$P_{ep}(T_{el}) = \left(\frac{k_{B}}{e}\right)^{2} \frac{L^{2}T}{R_{N}} \left(T_{el}^{5} - T_{Bath}^{5}\right), \quad (2)$$

where $L$ is the length of the normal wire and $\Gamma \approx 5 \cdot 10^{8}$ K$^{-3}$m$^{-2}$ for Au. The parameter $\Gamma$ is related to the electron-phonon scattering rate: $\tau_{ep}^{-1} = \zeta(3)/2\zeta(5)\Delta T_{el}^{3}$, where $\zeta(n)$ is the Riemann Zeta-function. The calculated cooling power according to Eqs. (1) and (2) is plotted as a function of $T_{el}$ in the wire (dotted and dashed line) in the inset of Fig. 2. For simplicity we assume $Z = 0$. The solid line is the sum of both contributions and corresponds to the solid line in the main figure 2. Finite values of $Z$ lead to a shift of the solid lines to lower cooling power and to higher electron temperatures, respectively. At intermediate temperatures both contributions are of comparable magnitude, while the electron-phonon term wins at low temperature because of the exponential cut-off of the N/S interface term and at high temperature because of the strong $T_{el}^{3}$-increase of the electron phonon term. In our geometry where the area of the N/S interface is tiny ($200 \times 200$ nm$^2$), the N/S interface term is much smaller than in the related experiment on Nb/Al/Nb junctions by Jehl et al. who used subtractive structuring of a Nb/Al bilayer. This may be the reason, why heating effects appear to be negligible in the latter experiment.

IV. AL/CU/AL - JUNCTIONS

It is now very interesting to look at samples, in which $L_{o}(\epsilon)$ remains larger than the wire length. To avoid inelastic scattering, it is necessary to keep $T_{el}$ below $\approx 1$ K in the voltage range $V \leq 2\Delta$. In a second set of experiments we replaced Nb by Al, having a much smaller gap $\Delta_{Al}$. As a consequence, the energies acquired in the MAR cycle are much lower and we expect to enter the regime of coherent Andreev reflection. For the normal wire we used both Au and Cu, where for Cu we measured a longer phase coherence length of 1.35 $\mu$m at 1.3 K than for Au. Figure 4 shows the resistance vs. temperature of a series of $16 \times 1 \mu$m long Al/Cu/Al junctions. When lowering $T$ the resistance sharply drops at the transition of the reservoirs $\approx 1.25$ K and then continuously vanishes as the proximity effect drives the Cu wire into a superconducting state. This sample shows zero resistance at the lowest $T$ since the Thouless energy $E_{c} = hD/L^{2} \approx 5\mu$eV and the normal state conductance are larger compared to the Nb/Au/Nb junctions. According to the theory by Wilhelm et al. the critical current $I_{c}(T)$ reads in the limit $k_{B}T \gg E_{c}$:

$$I_{c}(T) = \frac{3.0 \mu V/K}{R_{N} \sqrt{T_{0}}} T^{3/2} \exp \left(-\sqrt{T/T_{0}}\right), \quad (3)$$

where $T_{0} = E_{c}/2\pi k_{B}$.

The inset of Fig. 4 displays the measured current-voltage (IV) characteristics of the same sample. The turning point of the IV curves indicates $I_{c}(350\text{ mK}) \approx 300$ nA. With our sample parameters we estimate from
Eq. 3 a critical current $I_c (350 \text{ mK}) \simeq 510 \text{ nA}$. This estimate is reasonably close to the measured values. On the other hand, we observe a substantial broadening of the transition such that the zero voltage state is reached only for currents $\lesssim 80 \text{nA}$.

At finite temperatures a certain intrinsic broadening of the IV curves is expected by virtue of thermally activated phase slips which is usually described within the RSJ model. Our SNS junctions are self shunted with $R_N$ as the shunt resistance. We observe a broadening which is much stronger than expected from the RSJ model. This is illustrated by an RSJ fit using $I_c = 270 \text{nA}$ and $T = 350 \text{ mK}$ which is represented by the thick solid line in the inset of Fig. 4. In principle such an enhanced broadening can be caused by external electromagnetic interference. At high frequencies this source of broadening is suppressed by our rf filtering at room temperature and the sample stage. At lower frequencies we have checked, that the highest spikes in the frequency spectrum correspond to current noise below 1 nA/$\sqrt{\text{Hz}}$, which is much lower than the critical current at 350 mK. We are therefore confident that there is an intrinsic origin of the broadening of the IV curves. At voltages $V \gtrsim 5 \text{\mu V}$ the measured currents become larger than the fit. This is caused by the excess current induced by the Andreev reflection (see the discussion below).

Being made for tunnel junctions, a failure of the RSJ for long SNS junctions is not too surprising since it takes into account only the phase degree of freedom of the pair. An additional contribution to the shunt resistance is caused by the excess current induced by the Andreev reflection (see the discussion below).

The minimum value of $F(x)$ in SNS junctions is a minimum of $|F(x)|$ at the center of the N-wire. The minimum value of $|F(x)|$ at this ‘weak spot’ strongly depends on the ratio $L/L_T$ which is reflected in the temperature dependence of $I_c$ at temperatures $k_B T \ll \Delta$ described by Eq. 3. We believe that the enhanced rounding of the IV curves is related to the presence of the weak spot in the N-wire, which greatly facilitates phase slips in long SNS junctions. Correspondingly, also the shape of $R(T)$ cannot be fitted with the RSJ formulas, since the temperature dependence of $I_c$ is superimposed on that of the thermal activation process. In particular, $R(T)$ does not follow a simple Arrhenius law. Broadened transitions induced by phase slip processes also occur in microbridges and long filaments made from homogeneous superconductors. The latter examples differ from SNS junctions in that the main temperature dependence comes from $\Delta(T)$, which is not important at temperatures $T \ll T_c$.

The $dI/dV$ curves of the same sample but in a larger voltage range are presented for various temperatures in Fig. 5. Besides the supercurrent at $V = 0$ we find a considerable conductance enhancement for $V < 2\Delta$. In addition, conductance peaks close to $V = 2\Delta_{\text{Li}}/ne$ are present, which we attribute to coherent MAR cycles. The peaks are rather broad and the $n = 3$ peak appears even to be split. The inset in Fig. 5 displays the temperature dependence of the $2\Delta/e$ (i.e. $n = 1$) peak. The peak voltages nicely match the BCS curve with a slightly reduced gap. Being governed by $L_q(T)$ the amplitude
of the MAR features shows a relatively weak temperature dependence. This is in contrast to the supercurrent which strongly varies with temperature as expected from the exponential dependence of the Josephson coupling on $L_f$. Nearly identical observations have been made on Al/Au/Al junctions.

In order to further check that the peaks in $dI/dV$ are indeed related to the gap energy we measured another sample with different wire resistance. The critical current of the more resistive sample ($R_N = 34\, \Omega$) was substantially smaller but the peak voltages remained unaffected as demonstrated by the open symbols in the inset in Fig. 3. The value of the gap $\Delta(T = 0) = 163\, \mu$eV extracted from the $2\Delta$-peaks (see the inset in Fig. 3), is slightly reduced with respect to the bulk value of $186\, \mu$eV. Earlier experiments on conventional Nb/Nb point contacts have shown a significant suppression of the order parameter at the $n = 1$ peak which was attributed to a reduction of $\Delta$ by the relatively high currents which are required to generate the voltage $2\Delta$.

Another important quantity is the excess current $I_{exc} = (IV) - V/R_N$, i.e., the enhancement of the IV-characteristic above the ohmic straight line. $I_{exc}$ quantifies the integrated proximity correction to $dI/dV$ and saturates at large bias voltages $eV > 2\Delta$, where the Andreev reflection is suppressed. For superconducting point contacts with $E_c \gg \Delta$, $I_{exc}$ is predicted to be $I_{exc} = (\pi^2/4 - 1)\Delta/eR_N \approx 11\, \mu$A[3]. In the opposite limit of long diffusive junctions with $E_c \ll \Delta$, $I_{exc}$ is suppressed with increasing length as $1/L$ and amounts to $I_{exc} = 0.82\Delta/eR_N\xi^*/\Delta \approx 2.9\, \mu$A where $\xi^* = \sqrt{\hbar D_s/\Delta}$ and $D_s \approx 400\, \text{cm}^2/\text{s}$. When integrating the $dI/dV$ curves in Fig. 3 we find an asymptotic value of $I_{exc} \approx 3.5\, \mu$A, which is in acceptable agreement with the theoretical value obtained in the diffusive limit. The excess current is another feature, which is not contained in the RSJ model.

In the case of coherent MAR it is interesting to check for the existence of multiple charge $q^* = 2\Delta/V$ transferred during the MAR cycle, which should result in an enhanced shot noise $S_I = 2q^*I$ at low voltages[4]. A first indication for such an effect was seen in NbN based pinhole junctions[3]. In Fig. 3 we present noise data for the same sample as in Figs. 2 and 3. We indeed find a huge peak in $S_V$ at very low voltages around 3-4 $\mu$V $\approx 0.02\Delta/e$ which vanishes at elevated temperatures together with the supercurrent.

The noise enhancement appears in the strongly nonlinear part of $I(V)$ (inset in Fig. 3). The measured noise is frequency independent between 100 and 400 kHz (see inset in Fig. 3). The nonmonotonic dependence of $S_V$ on $V$ is in strong contrast to the $2\mu$m long Nb/Au sample of Fig. 3, which shows no supercurrent at our lowest temperatures and where $S_V$ is monotonically rising with $V$. At higher voltages we find an enhancement of $S_V$ for superconducting reservoirs (full circles) with respect to normal reservoirs (open triangles). In the voltage regime $V > \Delta$ the noise enhancement is most likely caused by heating similar to the Nb/Au case discussed above, where the heating is even more pronounced because of the larger gap of the Nb.

V. DISCUSSION

One possible origin of the low voltage noise peak are temporal fluctuations of the critical current as previously observed in grain boundary junctions made from high temperature superconductors[3]. Such critical current fluctuations may be caused by the motion of localized defects close to the junction and should result in a $1/f$ - like frequency dependence of the voltage noise close to $I_c$ as well as of the normal state resistance $R_N$. The latter would result in a parabolic increase of $S_V$ for $I > I_c$ which is absent in Fig. 3. At our typical measuring frequencies $f > 100\, \text{kHz}$ the measured peak height is independent of $f$ (see the inset in Fig. 3). For $f < 100\, \text{kHz}$ we observe a small increase of the peak amplitude which is currently not understood, but certainly inconsistent.
with a $1/f$ law. Hence, $1/f$ noise can be ruled out as the origin of the low voltage noise peak.

Earlier experiments on shunted tunnel junctions have also revealed an increase of the noise at low voltages. This effect has been predicted to arise as a consequence of Johnson-Nyquist noise of the shunt resistor. Fluctuations at high frequencies are mixed down to low frequencies by the highly nonlinear $IV$ characteristics of the junction. Good agreement with the experiment has been found for both the noise rounding of the $IV$ curves and the excess noise. When calculating the noise according to the RSJ model using the measured $dI/dV$ and $I_c(T)$ in the low voltage region for the temperatures shown in Fig. we find a peak with an amplitude of $35$ pV$^2$/Ω at $350$ mK which is about $20$ times smaller than the measured noise peak.

We believe that the noise peak is related to a strongly fluctuating supercurrent at the onset of finite voltage. As discussed already in the context of the $IV$ curves in Fig. temporal fluctuations of $|F(x)|$ can be thermally excited at the weak spot in the center of the N wire. These lead to large fluctuations of the supercurrent, and consequently to both large noise and unusually broad $IV$ curves. The minimum in $|F(x)|$ is the specific feature of junctions longer than $L_T$ and is not contained in the treatment of Refs.

The thermally activated fluctuations of the supercurrent have to be distinguished from the fluctuations of the critical current discussed above. The latter correspond to fluctuations of the activation energy with a $1/f$ spectrum, which are negligible at the time scale of µs, where our noise measurements are usually performed.

Independent support of this interpretation is provided by the recent observation of Thomas et al. who found a similar thermally activated rounding of the $IV$ characteristics in InAs-based SNS junctions. Their samples are also in the regime $L > L_T$ and the measured activation energy is typically two orders of magnitude smaller than expected from the RSJ model. In our samples, $R(T)$ is also broader than expected from the RSJ model (see the inset in Fig. ). Thomas et al. suggest that the rounding of the $IV$ characteristics may be caused by an additional current noise, which is much larger than the Johnson noise of the device. Our work provides direct experimental evidence for such an enhanced noise at the onset of Josephson coupling in “long” ($L > L_T$) SNS contacts.

In contrast to the supercurrent the MAR induced sub-harmonic gap structure in $dI/dV$ is much less temperature dependent (see Fig. ). The most striking signature of higher order MARs would be the presence of shot noise of multiple charge quanta. In order to check for MAR induced low voltage noise we have to look at higher temperatures where the supercurrent and its corresponding noise peak are suppressed. In Fig. we plot the effective charge $q^* e = S_I/2eI$ vs. $1/V$. At low temperatures $T \lesssim 500$ mK the noise peak in $S_V$ is reflected also in $q^*$. Remarkably, the noise raises again for even lower voltages, instead of dropping to zero as expected if the supercurrent noise is the only noise source. At higher temperatures $T \gtrsim 500$ mK the peak associated to supercurrent fluctuations vanishes, but we still find a noise signal which rises roughly linear with $1/V$. At the lowest voltages all curves (dashed lines) seem to merge into a straight line with a slope only slightly lower than \( \approx 0.3 \cdot 2\Delta \) as predicted by the theory for the diffusive regime (solid line). The theory by Naveh and Averin for the MAR noise considers very short junctions with $E_c \gg \Delta$. In our long SNS junctions $E_c \ll \Delta$ is the smallest energy scale. To our knowledge, the shot noise has not yet been calculated for this case.

![FIG. 7. Effective multiple charge $q^* = S_I/2eI$ as a function of $1/V$ corresponding to the data in Fig. 6. The solid line indicates the theoretical estimate for $q^*$ for the diffusive case. The dashed lines are a guide to the eye. The error bars indicate the uncertainty due to the subtraction of the background noise.](image)

The measured effective charge ranges up to $100 \, e$, which is surprisingly large since the coherence of the MAR cycle is expected to be cut off by inelastic scattering in our samples after a few Andreev reflections. From this point of view, it is already surprising that we find up to four MAR peaks in $dI/dV$. This raises the question whether phase coherence over $n \times L$ ($n$ is the number of Andreev reflections) is required or only over 1 or $2 \times L$. Although the magnitude and the functional dependence of the low voltage noise in Fig. are compatible with the existence of multiple charges, we cannot exclude other possibilities. Further experiments are required to separate the contributions from the supercurrent noise and the possible shot noise of multiple charge quanta.

VI. CONCLUSIONS

By means of noise measurements we have shown that multiple Andreev reflections in a normal metal wire sandwiched between two superconductors lead to substantial electron heating in the wire. The strength of this heating effect depends on the size of the gap in the supercon-
ductors. For Nb with a large gap the effective electron temperature raises already for small currents up to several K, which leads to a suppression of coherent multiple Andreev reflection. For the smaller gap superconductor Al the heating is less pronounced and phase sensitive effects such as subharmonic gap structure become visible. With the onset of a proximity induced supercurrent through the normal wire a sharp noise peak appears at low voltages, which we attribute to thermally induced fluctuations of the supercurrent. When the supercurrent is suppressed at moderately elevated temperatures an additive noise contribution remains at low voltages, which suggests the existence of multiple charge quanta with charge much larger than $e$.

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