Dissipation and Supercurrent Fluctuations in a Diffusive Normal-Metal–Superconductor Ring

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A mesoscopic hybrid normal-metal–superconductor ring is characterized by a dense Andreev spectrum with a flux dependent minigap. To probe the dynamics of such a ring, we measure its linear response to a high frequency flux, in a wide frequency range, with a multimode superconducting resonator. We find that the current response contains, besides the well-known dissipationless Josephson contribution, a large dissipative component. At high frequency compared to the minigap and low temperature, we find that the dissipation is due to transitions across the minigap. In contrast, at lower frequency there is a range of temperature for which dissipation is caused predominantly by the relaxation of the Andreev states' population. This dissipative response, related via the fluctuation dissipation theorem to a nonintuitive zero frequency thermal noise of supercurrent, is characterized by a phase dependence dominated by its second harmonic, as predicted long ago but never observed thus far.

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Connecting a phase coherent nonsuperconducting conductor (N) to two superconductors (a S-N-S junction) gives rise to the formation of Andreev states (AS) which are coherent superpositions of electron and hole states confined in the N metal and carry the Josephson supercurrent [1]. They strongly depend on the phase difference φ between the two superconductors. The quasicontinuous Andreev spectrum of a diffusive metallic wire exhibits a phase modulated induced gap, the minigap, which closes at odd multiples of π [2–4]. Theoretically, it was predicted that, in contrast to tunnel Josephson junctions [5] and because of the smallness of this induced gap, S-N-S junctions should exhibit low frequency and low temperature supercurrent fluctuations at equilibrium [6]. According to the fluctuation dissipation theorem, such equilibrium fluctuations led to a dissipative current under an oscillating flux excitation [7,8] and was predicted long ago to be characterized by a phase dependence dominated by its second harmonic [9], π periodic, but not yet observed. In this Letter, we present one of the most conceptually simple and direct evidences of these supercurrent fluctuations: the investigation of the linear current response of a phase biased N-S ring over a wide frequency range. We identify two fundamental dissipation mechanisms, the microwave-induced transitions across the minigap and the energy relaxation of Andreev level populations. We focus on this second contribution which is found to be nearly π periodic. The extra cusps we find at odd multiples of π reflect the closing of the minigap. This characteristic phase dependence, which is precisely that of the low frequency thermal supercurrent noise, as well as the frequency dependence are in complete agreement with theoretical predictions [6,8,9]. These experiments could be generalized to any S-N-S junction or S constriction if they contain at least one well transmitted conduction channel, so that the Andreev levels are close enough at π. This shows that linear ac measurements, close to equilibrium, reveal physical properties that are not accessible by standard transport measurements dominated by nonlinear effects such as switching current and ac Josephson effect [10,11].

To probe the AS close to equilibrium, the control of the phase difference across the junction is essential and can be done in a N-S ring threaded by an Aharonov-Bohm flux. dc experiments in this configuration include the tunnel spectroscopy of the minigap [12] and the measurement of the flux dependent Josephson supercurrent using superconduting quantum interference device [13] or Hall probe magnetometry [14]. Beyond the equilibrium AS spectrum in a *static* magnetic flux, we investigate its dynamics [15], using a technique pioneered for mesoscopic normal rings [16]. We couple a N-S ring to a superconducting microresonator and phase bias it with a dc Aharonov-Bohm flux and a small oscillating flux $\delta\Phi_\omega$ at the resonator's eigenfrequencies ω. The linear current response δI_ω is characterized by the complex susceptibility $\chi(\omega) = \delta I_\omega/\delta\Phi_\omega = \chi'(\omega) + i\chi''(\omega) = i\omega Y(\omega)$, where $Y(\omega)$ is the N-S ring's admittance. This susceptibility is extracted from the variations of the resonator's eigenmodes (frequency and quality factor). The ring's dissipationless response is deduced from the periodic flux variations of χ', whereas the dissipation corresponds to χ''. A first experiment [15] found a large dissipative response as well as a nondissipative one that differed notably from the adiabatic susceptibility, the simple flux derivative of the ring's Josephson current (the inverse kinetic inductance). These results were partially explained by the theory of the proximity effect [8]. However, the shape of the flux dependences of χ did not vary with frequency (in the range explored), so that the different components of the ring's dynamical response could not be accessed. In particular, with the inelastic scattering rate much smaller than the lowest eigenfrequency, the dissipative response associated with the relaxation of Andreev states could not be detected. In the present experiment, we report on a N-S ring with an enhanced temperature dependent inelastic scattering rate $1/\tau_{\rm in}(T)$, thanks to a thin Pd layer at the N-S interface, between the normal gold mesoscopic wire and the superconducting niobium loop. The higher inelastic scattering rate, combined with a broader frequency and temperature range, leads to the identification of the two fundamental contributions to the supercurrent relaxation. At frequencies above the inelastic scattering rate, dissipation is due to microwave-induced excitations across the minigap. In the opposite regime of lower frequency, which could not be reached previously, dissipation is due to the thermal relaxation of Andreev level populations.

The experimental setup is shown in Fig. 1(a): the resonator consists of a double meander line etched out of a 1 μm thick niobium film sputtered onto a sapphire substrate. The N-S ring connects the two lines at one end of the resonator, turning it into a $\lambda/4$ line with a fundamental frequency of 190 MHz, and harmonics 380 MHz apart. A weak capacitive coupling to the microwave generator preserves the high quality factor of the resonances, which can reach 5×10^4 up to 10 GHz. The N-S ring is fabricated by electron beam lithography. The Au wire (4 μm long, 0.3 μm wide, and 50 nm thick) is first deposited by e-beam deposition of high purity gold. The S part is deposited in a second alignment step by sputtering of a Pd/Nb bilayer (6 nm Pd, 100 nm Nb). The resulting uncovered length of the Au wire is $L = 1$ μm, corresponding to the long junction limit $L \gg \xi_S$, the superconducting coherence length. The N-S ring is connected to the Nb resonator in a subsequent step, using ion-beam assisted deposition of a tungsten wire in a focused ion beam (FIB) microscope. This process creates a good superconducting contact between the resonator and the Pd/Nb part of the ring. The 6 nm thick Pd buffer layer ensures a good transparency at the N-S interface, as demonstrated by the amplitude of the critical current measured with dc transport measurements on control S-N-S junctions fabricated simultaneously [Fig. 1(c)]. It also enhances the inelastic scattering rate because of Pd's spin wave–like excitations (paramagnons) [17–19]. Considering that the phase coherence time extracted from weak localization measurements on a 6 nm thick Pd thin film [17] was of the order of 0.3 ± 0.1 ns at 1 K, which is longer than the estimated diffusion time $\tau_D = 0.1$ ns through the Au wire between the S contacts, we do not expect a reduction of the critical current as confirmed by measurements in the control samples.

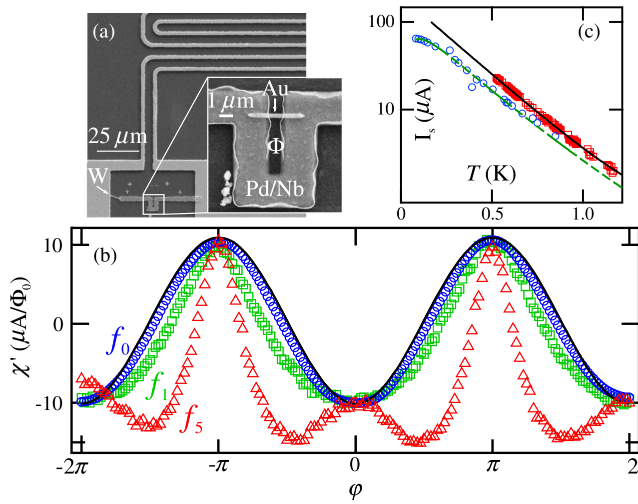

FIG. 1 (color online). (a) Inset: Scanning electron micrograph of the N-S ring fabricated to explore the dynamics of Andreev states. (Main panel) N-S ring is connected to a Nb multimode resonator (meander in the top) by W wires deposited by a FIB. (b) Flux dependence of χ' at several resonator eigenfrequencies ($f_0 = 190$ MHz, $f_1 = 560$ MHz, $f_5 = 2$ GHz) and $T = 1.2$ K. At f_0, χ' is barely distinguishable from a cosine (solid line). Whereas the amplitude $\delta\chi'_{\pi-0} = \chi'(\pi) - \chi'(0)$ is frequency independent, its temperature dependence is that of the switching current I_s. Note the appearance of a local maximum around $\varphi = 0$ at high frequency. (c) Temperature dependence of the switching current of the control sample I_s (circles) and of $(\Phi_0/2\pi) \times (\mathcal{L}/L_C^2)\delta\chi'_{\pi-0}$ measured in the N-S ring (squares) with their fits according to Ref. [11] (respectively, dashed and solid lines). Best fit yields $E_{\rm Th} \equiv 71 \pm 5$ mK, corresponding to 1.5 GHz for the N-S ring.

The quantities we measure are the variations with dc flux of the resonator's quality factor and eigenfrequencies $\delta Q(\Phi)$ and $\delta f(\Phi)$. They are simply related to the oscillating phase dependent part of the complex susceptibility, characterized by $\chi'(\varphi)$ and $\chi''(\varphi)$, where the superconducting phase φ is related to the flux threading the ring by $\varphi = -2\pi\Phi/\Phi_0$, where $\Phi_0 = h/2e$ is the superconducting flux quantum. The relation reads [15]

$$\frac{\delta f_n(\Phi)}{f_n} = -\frac{1}{2}\frac{L_C^2}{\mathcal{L}}\chi'(\varphi, f_n),$$
$$\delta\frac{1}{Q_n}(\Phi) = \frac{L_C^2}{\mathcal{L}}\chi''(\varphi, f_n). \tag{1}$$

The coupling inductance $L_C = 9 \pm 2$ pH is due to the S part of the N-S ring, and $\mathcal{L} = 0.3$ μH is the resonator's geometrical inductance. These expressions are valid at temperatures such that the kinetic inductance of the S-N-S junction is larger than the ring's geometrical inductance (outside this range screening of the applied flux, both dc and ac, needs to be considered [15]). This sets the lower limit to the temperature, so that experiments were conducted between 0.4 and 1.5 K. The frequencies probed ranged between 190 MHz and 3 GHz.

We find drastic variations of both the amplitude and the shape of χ' and χ'' as frequency and temperature are changed. At the lowest frequencies and highest temperatures investigated [see Figs. 1 and 2(a)], the dissipationless $\chi'(\varphi)$ is well described by a pure 2π-periodic cosine, as expected for the adiabatic susceptibility $\chi_J = \partial I_J/\partial\Phi$ of the Josephson current which is purely sinusoidal at these moderately high temperatures, much larger than E_{Th} [2,14]. As shown in Figs. 1(b) and 1(c), the amplitude $\delta\chi'_{\pi-0} = \chi'(\pi) - \chi'(0)$ perfectly reflects the expected, roughly exponential, decay of the Josephson critical current $I_J(T) = I_J(0)\exp(-k_BT/3.6E_{\mathrm{Th}})$ [11], that was also measured in the control wire, with E_{Th} the Thouless energy $\hbar/\tau_D$ and τ_D the diffusion time across the N wire.

We find for both samples $E_{\mathrm{Th}} = 71 \pm 5$ mK, which corresponds to $\tau_D \simeq 0.1$ ns and $E_g(0) \simeq 3E_{\mathrm{Th}} = 210$ mK for the maximum amplitude of the minigap at $\phi = 0$. In contrast, the dissipation, characterized by $\chi''(\varphi)$, is nearly π periodic [see Fig. 2(b)] at the highest temperatures investigated and acquires a strong 2π-periodic component at lower temperature. When increasing the frequency [Figs. 1(b), 2(a), and 2(c)], $\chi'(\varphi)$ contains additional harmonics, with peaks at odd multiples of π and moreover a local maximum at $\varphi = 0$, mod $[2\pi]$ for the highest temperatures. On the other hand, at 2 GHz [Figs. 2(c) and 2(d)] and low temperature, $\chi'(\varphi)$ and $\chi''(\varphi)$ have identical shapes, with peaks at π, mod $[2\pi]$, reflecting the underlying minigap that varies like $2E_g(0)|\cos(\varphi/2)|$.

In the following we exploit this complex evolution of $\chi(\varphi)$ with frequency and temperature to extract the different mechanisms at work in the dynamics of AS. To this

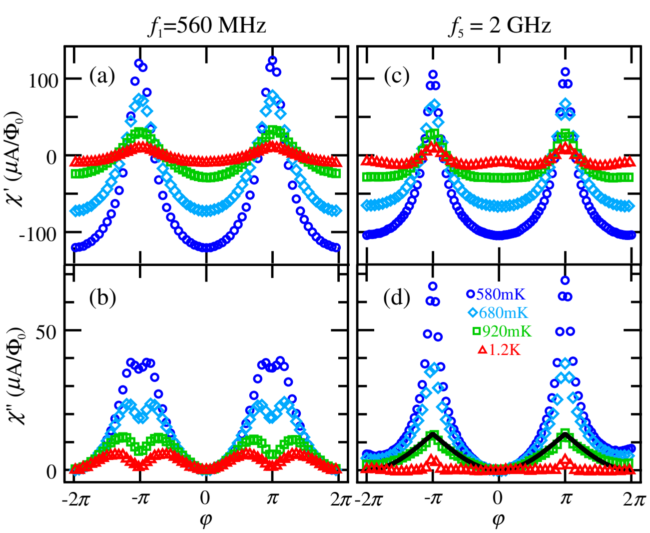

FIG. 2 (color online). Evolution of χ' [top, (a) and (c)] and χ'' [bottom, (b) and (d)] phase dependence with temperature at $f_1 = 560$ MHz $< E_{\mathrm{Th}}/h$ (left) and $f_5 = 2$ GHz $\gtrsim E_{\mathrm{Th}}/h$ (right). $\chi'(\varphi = \pi)$ and $\chi''(\varphi = \pi)$ increase with decreasing temperature. $\chi'(\varphi)$ and $\chi''(\varphi)$ strongly differ at low frequency and high temperature, whereas they are similar with a shape reminiscent of the minigap at high frequency. The solid line in (d) is proportional to opposite of the minigap dependence $|\cos(\varphi/2)|$. Curves have been shifted so that $\chi'(\varphi = \pi) - \chi'(\varphi = 0) = 0$ and $\chi''(\varphi = 0) = 0$.

end, we make use of theoretical predictions [8] based on Usadel equations and recent numerical simulations [20] inspired by the analysis of the ac response of normal mesocopic rings [21–23]. The response function of a N-S ring has been shown to contain three contributions: $\chi = \chi_J + \chi_D + \chi_{ND}$. The adiabatic, zero frequency, Josephson contribution χ_J is purely real and is the derivative of the Josephson current $\partial I_J/\partial\Phi$. The second contribution, the diagonal susceptibility χ_D, is the first nonadiabatic, frequency dependent contribution. It describes the Debye-like relaxation of the (phase dependent) thermal populations $f_n(\varphi)$ of the Andreev states, with a typical inelastic relaxation time τ_{in} according to the simple model proposed for the dynamics of persistent currents in normal rings [21–23]:

$$\chi_D = \sum_n i_n \frac{\partial f_n}{\partial\Phi}\frac{i\omega}{1/\tau_{\mathrm{in}} - i\omega} = -\sum_n i_n^2 \frac{\partial f_n}{\partial\epsilon_n}\frac{i\omega}{1/\tau_{\mathrm{in}} - i\omega}, \tag{2}$$

where the square of $i_n = -\partial\epsilon_n/\partial\Phi$, the current carried by the nth Andreev level of energy $\epsilon_n(\Phi)$, appears. Finally, the nondiagonal contribution χ_{ND} describes quasiresonant microwave-induced transitions between two Andreev levels, involving (in contrast with χ_D) nondiagonal matrix elements of the current operator [20,24]. The contribution χ''_{ND} to the phase dependent susceptibility dominates when $\omega \geq 1/\tau_D \gg 1/\tau_{\mathrm{in}}$, as in Fig. 2(d). χ' and χ'' then have similar shapes which follow approximately the opposite of the minigap with peaks at π and a $-|\cos(\varphi/2)|$ dependence [20]. This high frequency regime was the only one accessed in the previous experiments on Au wires directly connected to W superconducting wires. In those experiments the energy relaxation time, limited by electron-electron interactions, of the order of 0.1 μs, was very long due to the superconducing contacts [15,25]. Therefore, those measurements were always in the regime $\omega\tau_{\mathrm{in}} \gg 1$, where χ''_D is negligible [Eq. (2)]. In contrast, the Pd layer beneath the Nb contacts in the present samples considerably reduces the inelastic scattering time, leading to a substantial contribution of χ''_D for the resonator's first five eigenfrequencies. We now focus on this contribution analyzed in Figs. 3 and 4.

We first present the predicted flux dependence of χ_D, given by the function F,

$$F(\Phi, T) = \sum_n i_n \frac{\partial f_n}{\partial\Phi} = -\sum_n i_n^2 \frac{\partial f_n}{\partial\epsilon_n}, \tag{3}$$

which reads in the continuous spectrum limit $F(\Phi, T) = \int d\epsilon J_S^2(\Phi, \epsilon)/[4k_BT\rho(\epsilon)\cosh^2(\epsilon/2k_BT)]$. Here, J_S and ρ are, respectively, the spectral current and the density of states of the S-N-S junction. This function was introduced by Lempitsky [9] to describe the $I(V)$ characteristics of S-N-S junctions and was calculated numerically using Usadel equations by Virtanen *et al.* [8]. At large temperature compared to E_{Th}, $F(\Phi, T)$ can be approximated by the

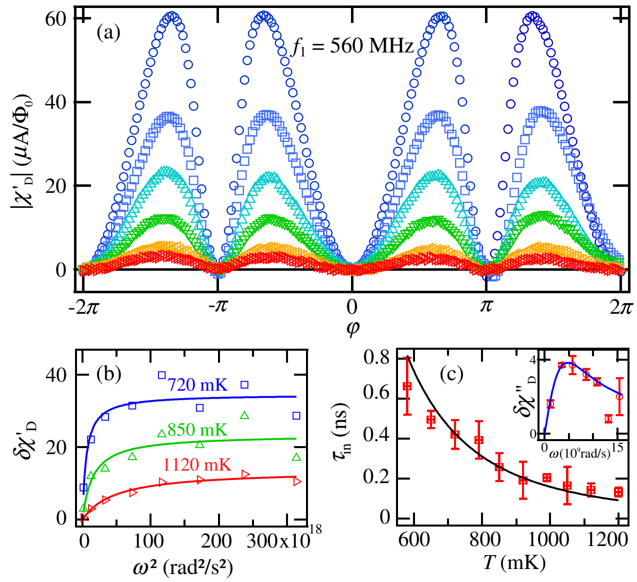

FIG. 3 (color online). (a) Experimental phase dependence of χ'_D at $f_1 = 560$ MHz and (from top to bottom) $T = 580$, 650, 720, 820, 1000, 1200 mK. In agreement with theory [8], the position of the maximum slightly shifts to lower values with decreasing temperature. (b) Frequency dependence of $\delta\chi'_D$: the maximum of $-\chi'_D(\varphi)$, at different temperatures (triangles), compared to the theoretical prediction from Eq. (3). (c) Temperature dependence of extracted τ_{in} (see text). Solid line is a T^{-3} law. Inset: Frequency dependence of $\delta\chi''_D$ at 1.15 K (circles) compared to the theoretical prediction from Eq. (2).

following analytical form: $F_U(\varphi, T) \propto (\{-\pi + (\pi + \varphi) \times [2\pi]\}) \sin(\varphi) - |\sin(\varphi)| \sin^2(\varphi/2)/\pi$. It is dominated by its second harmonics with in addition a sharp linear singularity at odd multiples of π (see Fig. 4). This is due to the dominant contribution of Andreev levels close to the minigap whose flux dependence is singular like in a highly

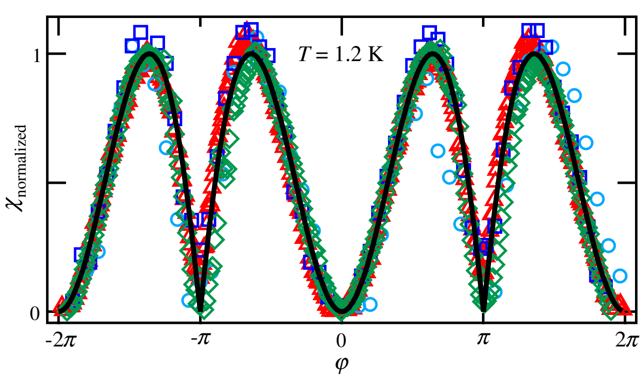

FIG. 4 (color online). Normalized experimental phase dependences at $T = 1.2$ K (open symbols) compared to F_U (solid). Data are χ'' (squares), $|\chi'_D| = \chi_J - \chi'$ (circles) at $f_1 = 560$ MHz and χ'' (triangles), $|\chi'_D| = \chi_J - \chi'$ (diamonds) at $f_3 = 1.35$ GHz. At high temperature and for $f \lesssim E_{\text{Th}}/h$, experimental $\chi_J - \chi'$ and χ'' are found to be in very good agreement with theoretical prediction of Usadel equations F_U.

transmitting superconducting single channel point contact [6]. We now show that the particular flux dependence of the function F_U can explain the experimental data of Figs. 1 and 2. We first follow the frequency dependence of the amplitude of $\chi'_D(\varphi) = \chi'(\varphi) - \chi_J(\varphi)$ at fixed temperature and check that the shape of $\chi'_D(\varphi)$ does not change with frequency and is the same as that of χ''_D, as predicted for the temperature and frequency regime where the contribution of χ''_{ND} can be neglected. As shown on Fig. 3(b), it is then possible to fit the frequency dependence of the amplitude of $\chi'_D(\varphi)$ by the expected $(\omega\tau_{in})^2/[1 + (\omega\tau_{in})^2]$ and determine the characteristic time τ_{in} for several temperatures according to Eq. (2). We find values of τ_{in} varying between 1 and 0.2 ns. This last value measured at 1 K is of the same order as the value deduced from weak localization measurements in Pd thin films [17] at the same temperature. The T^{-3} decrease of $\tau_{in}(T)$ could be attributed to paramagnons since they cause the largest inelastic scattering at low temperature in Pd (a metal close to a ferromagnetic transition). It is also interesting to note that our results can be described by a single inelastic time, independent of φ, whereas a phase dependent τ_{in} is expected for electron phonon collisions in S-N-S junctions [26]. This is probably due to the fact that temperature is larger than the amplitude of the minigap in our case.

A similar analysis can be done on χ''. The quality of the calibration is, however, not as good as on χ'. Moreover, we still lack a good analytical prediction for $\chi_{ND}(\varphi)$, which gives a large contribution to $\chi''(\varphi)$ at low temperature and high frequency. We have overcome this difficulty by subtracting for frequencies larger than 1.7 GHz the flux dependence of χ''_{ND} estimated from the high frequency data (2.8 GHz). The resulting amplitude $\delta\chi''_D(\omega)$ agrees with the expected frequency dependence in $\omega\tau_{in}/[1 + (\omega\tau_{in})^2]$ as shown in the inset of Fig. 3(c). One can also compare the independently measured flux dependences of $\chi' - \chi_J$ and χ'' with theoretical predictions from the Usadel equations, $F_U(\Phi, T)$. This is done in Fig. 4 for several frequencies, and a good agreement is found.

With this set of experiments, we have thus shown that the frequency and temperature dependences of the response function of N-S rings in a time dependent flux are consistent with a simple Debye relaxation model of the population of the Andreev levels. Using fluctuation dissipation theorem one can estimate the related thermodynamic current noise as

$$S_I(\omega) = \frac{2}{\pi} \frac{k_B T \chi''_D(\omega)}{\omega} = \frac{2}{\pi} k_B T \sum_n i_n^2(\varphi) \frac{\partial f_n}{\partial \epsilon_n} \left[\frac{\tau_{in}}{1 + (\omega\tau_{in})^2}\right]. \quad (4)$$

Interestingly, the expression of this noise becomes particularly simple when temperature is much larger than the minigap, so that $(\partial f_n/\partial \epsilon_n)$ is given by $1/k_B T$. One then finds that the frequency integrated current noise is just

$\delta I^2 = \sum_n i_n^2$, which corresponds to independent current fluctuations for each Andreev level.

The measurement of the current linear response of a N-S ring to a high frequency flux thus reveals two fundamental mechanisms contributing to dissipation at finite frequency. One of them, predominant at high frequency and low temperature, describes the physics of microwave-induced transitions above the minigap. We have clearly identified and characterized the second cause of dissipation, the thermal relaxation of the populations of the Andreev states. It is described by an inelastic rate which is extremely sensitive to the nature of the N-S interface. This dissipative response is directly related to the low frequency thermal noise of the Josephson current, with a flux dependence proportional to the average square of the spectral (or single level) current and can be precisely described by theoretical predictions. The type of experiments presented here is uniquely suited to investigate more exotic systems, for instance, with the normal diffusive wire replaced by a ballistic wire, leading to a discrete Andreev spectrum known to be extremely sensitive to spin orbit interactions [27].

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