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Microwave Admittance of Gold-Palladium Nanowires with Proximity-Induced Superconductivity

Russell E. Lake,* Joonas Govenius, Roope Kokkoniemi, Kuan Yen Tan, Matti Partanen, Pauli Virtanen, and Mikko Möttönen

Quantitative electrical admittance measurements of diffusive superconductor-normal-metal-superconductor (SNS) junctions at gigahertz frequencies and millikelvin temperatures are reported. The gold-palladium-based SNS junctions are arranged into a chain of superconducting quantum interference devices. The chain is coupled strongly to a multimode microwave resonator with a mode spacing of approximately 0.6 GHz. By measuring the resonance frequencies and quality factors of the resonator modes, the dissipative and reactive parts of the admittance of the chain are extracted. The phase and temperature dependence of the admittance near 1 GHz are compared with theory based on the time-dependent Usadel equations. This comparison allows the identification of important discrepancies between theory and experiment that are not resolved by including inelastic scattering or elastic spin-flip scattering in the theory.

1. Introduction

The transport of direct current (dc) between two superconductors (S) separated by a diffusive normal-metal (N) link is in general well understood both theoretically and experimentally.^[1–6] At low temperatures and currents, Andreev reflection^[7] leads to the formation of a gap in the density of quasiparticle states in N and allows a dissipationless supercurrent to flow. This gap has been directly observed in tunnel spectroscopy



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measurements.^[8,9] Shapiro steps, supercurrent enhancement, and other non-equilibrium effects under intense microwave irradiation have also been extensively studied.^[1,10–16]

In contrast, the near-equilibrium response of superconductor–normalmetal–superconductor (SNS) junctions to weak microwave radiation has become an active area of investigation only recently.^[17–21] While the adiabatic contribution to the kinetic inductance can be calculated from the dc current–phase relation, at high frequencies both reactive and dissipative contributions arise also from other mechanisms, such as driven transitions between quasiparticle states and oscillation of the Andreev level

populations.^[18] Surprisingly, however, little experimental data has been published on the topic thus far.^[22,23] The parameter regimes and materials studied in the published experiments are very sparse, hence limiting the extent to which theoretical predictions^[17–21] can be tested. In practice, data on the effective inductance and losses also expedites the process of designing high-frequency SNS-junction-based circuits, such as the SNS nanobolometer.^[24,25]

Previous experimental studies^[22,23] have probed flux- and temperature-dependent changes in the linear microwave response of a superconducting ring with a gold normal-metal inclusion. The superconducting ring consisted of ion-beamdeposited tungsten in the first experiments,^[22] and sputterdeposited Nb with a thin Pd layer at the SN interface in the later experiments.^[23] A single SNS ring was biased with a dc magnetic flux and coupled weakly to a multi-mode microwave resonator. By measuring flux-dependent shifts in the quality factors and resonance frequencies, the authors determined how the complex-valued electrical susceptibility χ changes.^[22,23] The change in χ , as a function of flux and temperature, was reported to be in excellent agreement with theoretical predictions based on Usadel equations and numerical simulations. However, the implicit offsets in both the real and imaginary parts of the reported susceptibility prevent a comparison to the theoretically predicted absolute values of $\operatorname{Re}[\chi]$ and $\operatorname{Im}[\chi]$. They also prevent the accurate prediction of the effective inductance ($\operatorname{Re}[\chi]^{-1}$) and the loss tangent $(\text{Im}[\chi]/\text{Re}[\chi])$, which are the key quantities for practical high-frequency applications of SNS junctions.

In this article, we present measurements of the SNS junction admittance $Z^{-1}(\omega) = \chi/i\omega$ for gold-palladium based junctions at angular frequencies ω of order $2\pi \times 1$ GHz. We use a chain of

 $\begin{array}{c} \text{materials} \\ \text{ww.advelectronicmat.de} \\ \begin{array}{c} \text{a)} & \text{resonator} & \text{termination} \\ \hline \\ \text{FL} & \hline \\ \text{FL} & \hline \\ \text{c} & f_0 \approx 635 \text{ MHz} & C_i & R & C_{gnd} \\ \hline \\ \text{fr} & f_1 \approx 10 \text{ cm} & \hline \\ \text{c} & f_1 \approx 10$

Figure 1. a) On-chip circuit layout: feedline (FL) between ports 1 and 2, transmission line part of the resonator, and termination. The resonances are externally damped by the capacitive coupling (C_c) to the feedline, and internally damped by the capacitively coupled SQUID chain. See Table 1 for further details on parameter values. b) Schematic detail of the device under test at the termination: 20 SNS SQUIDs in series. c) Micrograph of a single SQUID from a sample fabricated identically to Sample 2. The wide Au–Pd extensions (light-colored regions) are heat sinks.

SNS superconducting quantum interference devices (SQUIDs) with a strong capacitive coupling to a multimode microwave resonator with a typical mode spacing of 0.6 GHz. Each chain consists of 20 SNS SQUIDs in series. The strong coupling and the large number of SQUIDs lead to significant changes in the frequencies and quality factors of the resonator modes, which allow determining Z^{-1} without an offset. The absence of an offset enables us to show that the Usadel-equation-based theory we consider cannot simultaneously explain the observed real and imaginary parts of Z^{-1} .

2. Samples

We study the linear electrical response of SNS junctions at millikelvin temperatures in two samples: Sample 1 and 2. The gold-palladium nanowires used as the normal-metal are deposited simultaneously in the same fabrication steps as for our recent nanobolometer circuits.^[25] We determine the Au:Pd atomic ratio of the alloy to be approximately 3:2 using energydispersive X-ray spectroscopy (see Experimental Section). The nominal junction length *l* is 300 nm and the nominal cross-sectional area is 30 nm \times 120 nm. The normal-state resistance of a single junction $R_{\rm N} = 15 \ \Omega$ is estimated based on four-wire dc measurements of reference samples. From the reference measurements, we also estimate the upper bound for the contact resistance $R_{\rm B}$ to be approximately 1 Ω . We only give an upper bound for the contact resistance because of the uncertainties introduced by the crossed-wire geometry^[26] of the reference samples. Reference samples are similar to those in Figure 1b in Reference [25].

The key parameter determining the strength of the proximity-induced superconductivity is the Thouless energy $E_{\rm T} = \hbar D l^{-2}$, where $D \approx 22 \text{ cm}^2 \text{ s}^{-1}$ is the diffusion constant. We obtain the diffusion constant by measuring the resistivity of the nanowires and scaling the result according to literature values for the proportionality $D \propto \rho^{-1}$.^[27] The result we obtain is approximately 30% higher than that in Reference [27]. In our samples, the Thouless energy is $E_{\rm T} \approx k_{\rm B} \times 190 \text{ mK} \approx$ $h \times 3.9 \text{ GHz}$, where $h = 2\pi\hbar$ is the Planck constant and $k_{\rm B}$ is the Boltzmann constant. The superconducting sections are 100-nm-thick aluminum, which implies that the energy gap Δ in the superconductors is much larger than the Thouless energy ($\Delta/E_{\rm T} \approx 13$). Both S and N parts are fabricated using electron beam lithography and evaporation. Further fabrication details are reported in the Experimental Section.

The SNS junctions are arranged into a chain of SQUIDs, as shown in **Figure 1**. Each SQUID loop has a relatively small area of 20 μ m² in order to minimize sensitivity to external magnetic field noise. In addition, we measured Sample 2 in a double-layer magnetic shield. The geometric inductance of each loop is small ($L_{\rm G} < 10$ pH) compared to the effective inductance of the SQUID, as verified by the results below. A dc magnetic flux bias is applied by an external coil that provides a uniform flux bias Φ for each SQUID in the 100- μ m-long chain. Assuming identical junctions, the field biases each SNS junction at a phase difference $\pi(\Phi - m\Phi_0)/\Phi_0$ at dc, where Φ_0 is the magnetic flux quantum h/2e and *m* is the integer that minimizes $|\Phi - m\Phi_0|$. We note that the flux bias we label as $\Phi = 0$ may be offset from the actual zero flux condition by an integer multiple of Φ_0 .

The SQUID chains in the two samples are nominally identical, except for the addition of heat sinks to Sample 2 (see Figure 1c). The heat sinks are designed to reduce the hot-electron effect,^[28] i.e., the increase of the quasiparticle temperature above the phonon bath temperature $T_{\rm b}$ that we measure. For each SNS junction, the heatsink consists of two large (0.5 µm³) reservoirs of gold-palladium that are thermally strongly coupled to the junction.

In addition to the SQUID chain, the chip contains a transmission line resonator of length 10 cm (see Figure 1a and **Table 1**, and Figures S1 and S2, Supporting Information). We characterize it by measuring control samples with an open

Table 1. Resonator and coupling capacitor parameters for Samples 1 and 2: the internal (external) load capacitance C_i (C_c), the transmission line resonator length l_n the fundamental frequency f_0 , and the resonator chacteristic impedance Z_0 shown in Figure 1. The last column emphasizes that Sample 2 includes additional large gold-palladium heat sink reservoirs for enhancing electron–phonon coupling.

Sample	C _C [pF]	C _i [pF]	l _r [mm]	<i>f</i> ₀ [MHz]	Ζ ₀ [Ω]	Heat sinks
1	0.15	14.5	97.2	637	39	No
2	0.44	15.2	96.5	633	39	Yes



termination, i.e., samples without the SQUID chain. From the control measurements, we extract the fundamental frequency of the transmission line resonator $f_0 \approx 635$ MHz, and confirm that the internal quality factor $Q_{i,n} > 10^4$ for the resonances we consider (n \approx 3). The latter implies that we can neglect the losses in the transmission line part of the resonator, and in the Al₂O₃ used as the dielectric material in the lumped element capacitors (C_C and C_i). This is valid because introducing the SQUID chain lowers $Q_{i,n}$ to the order of 10^2 , as observed below. We also deduce the characteristic impedance $Z_0 \approx 39 \Omega$ of the transmission line from the measured f_0 , the length of the resonator, and the design value for the inductance per unit length.

3. Measurement Scheme and Sample Characterization

We determine the admittance of the SQUID chain by embedding it as the termination of the transmission line microwave resonator, as illustrated in Figure 1a. We first determine the resonance frequency f_n and the internal quality factor $Q_{i,n}$ of each mode *n* by measuring the frequency-dependent transmission coefficient $S_{21}(f)$ through the feedline. By comparing f_n and $Q_{i,n}$ to values measured in control samples with no SQUID chains, we can determine the admittance of the SQUID chain at multiple frequencies. Specifically, we use a circuit model (Figure 1a) that allows extracting the admittance of the SQUID chain Z_{chain}^{-1} from the response of the combined resonator/SQUID-chain system. The admittance of each individual SNS junction is then given by 10 Z_{chain}^{-1} , assuming that the junctions are identical and that geometric inductance is negligible.

Figure 2 shows the normalized transmission through the feedline at frequencies near 1.4 GHz, probing the third (n = 3) mode in Sample 2. The normalization (defined precisely in the Experimental Section) removes all spurious features in the transmission data that do not depend on flux. This procedure reveals the oscillatory flux dependence of the resonance frequency f_n with a period we identify as Φ_0 . As the flux bias is increased away from integer multiples of Φ_0 , we measure a decrease in both the resonance frequency and the loaded quality factor $Q_{0,n}$. This behavior is more clearly visible in Figure 2b,c with individual slices of transmission data for $\Phi/\Phi_0 = 0$ and $\Phi/\Phi_0 = 0.3$. These changes in the resonance indicate that both the inductance and the losses are maximized in the SQUID chain near half-integer values of Φ/Φ_0 .

To extract f_n and $Q_{j,n}$ quantitatively, we fit the measured normalized transmission for the *n*th mode to the model^[29,30]

$$S_{21}(f) = 1 - \frac{\frac{Q_{0,n}}{Q_{C,n}} - 2iQ_{0,n}\frac{\delta f}{f_n}}{1 + 2iQ_{0,n}\frac{f - f_n}{f_n}}$$
(1)

where δf is a fit parameter that characterizes asymmetry, and the external quality factor $Q_{C,n}$ is governed by the coupling (C_C) to the feedline. From the obtained fit parameters $Q_{0,n}$ and $Q_{C,n}$, we compute the contribution of losses due to the SQUID chain as $Q_{1,n}^{-1} = Q_{0,n}^{-1} - Q_{C,n}^{-1}$. **Figure 3** shows the extracted values of f_n and $Q_{1,n}$ for frequencies up to 12 GHz ($n \approx 20$).



Frequency / GHz Frequency / GHz

Figure 2. a) Magnitude of the normalized transmission coefficient $|S_{21}|$ for the third resonator mode (n = 3) of Sample 2 at $T_b = 10$ mK as a function of frequency and flux-bias Φ . b) Traces from a) along $\Phi/\Phi_0 = 0$ and $\Phi/\Phi_0 = 0.3$ with fits (solid lines) to the model in Equation (1). c) Same as (b) but for the phase arg(S_{21}). The extracted resonance frequencies and internal quality factors are $f_3 = (1452.6 \pm 0.4)$ MHz; $Q_{i,3} = 127 \pm 8$ and $f_3 = (1411.1 \pm 0.4)$ MHz; $Q_{i,3} = 78 \pm 4$ for $\Phi/\Phi_0 = 0$ and $\Phi/\Phi_0 = 0.3$, respectively. The discontinuities in the background in a) are artifacts caused by the normalization procedure of S_{21} (see Experimental Section).

In the low-frequency and low-temperature regime, the SQUID chain behaves like an inductor, i.e., most of the admittance is reactive (Im[Z_{chain}^{-1}] >> Re[Z_{chain}^{-1}]) and Im[ωZ_{chain}^{-1}] varies slowly as a function of the angular frequency $\omega = 2\pi f$. Consequently, we parametrize the admittance Z_{chain}^{-1} as a parallel combination of a resistor and an inductor, such that $Z_{chain}^{-1} = R^{-1} + (i\omega L)^{-1}$. Figure 3 demonstrates that this is a good parametrization by showing qualitative agreement between the experimental data and predictions for the mode shifts and quality factors using a simplified model where *L* and *R* are constant.

Let us now discuss the extraction of the admittance Z_{chain}^{-1} from the measured f_n and $Q_{i,n}$ values. For an ideal transmission line resonator with open-circuit conditions at both ends $(C_{\rm C} = C_{\rm i} = 0)$, the *n*th mode is located at frequency nf_0 . In contrast, for the samples with the SQUID chains, the frequencydependent reactive, i.e., imaginary parts of the termination admittances $i\omega C_{\rm C}$ and $[(i\omega C_{\rm i})^{-1} + Z_{\rm chain}^{-1}]$ lead to the non-zero modeshift of Figure 3, that we use to determine $\rm{Im}[Z_{\rm chain}^{-1}]$. Similarly, the measured $Q_{i,n}$ gives information about the dissipative, i.e., real part of $Z_{\rm chain}^{-1}$.



Figure 3. Measured mode shift $f_n/f_0 - n$ (filled black circles) and internal quality factor $Q_{i,n}$ (filled red squares) for a) Sample 1 and b) Sample 2 at 10-mK phonon bath temperature and zero flux bias. For reference, the open markers show the theoretical prediction based on Equation (2) for a simplistic SQUID chain admittance $Z_{chain}^{-1} = R^{-1} + (i\omega L)^{-1}$ with a constant R equal to 350 Ω (500 Ω) and a constant L equal to 2.5 nH (5.0 nH) for Sample 1 (2). The dashed lines emphasize that the mode shift is zero for all harmonics of an ideal $\lambda/2$ resonator. Some f_n and Q_i values could not be experimentally extracted due to the presence of nearby parasitic resonances.

Quantitatively, we determine the SQUID chain admittance Z_{chain}^{-1} from $\omega_n = 2\pi f_n$ and $Q_{i,n}$ numerically solving the transcendental equation

$$i \tan\left[\frac{\omega_n}{2f_0}\left(1+\frac{i}{2Q_{i,n}}\right)+\arctan\left(Z_0\omega_nC_C\right)\right] = -\frac{Z_0}{\left(i\omega_nC_i\right)^{-1}+Z_{\text{chain}}\left(\omega_n\right)}$$
(2)

using the parameters given in Table 1. We derive this equation from the circuit model shown in Figure 1, assuming that $Q_{i,n}$ is dominated by losses in the SQUID chain.

Table 2 shows the L and R extracted for two example resonances near 1 GHz. The reported values provide an important reference for designing high-frequency devices based

Table 2. SQUID chain admittance parameters $R^{-1} + (i\omega L)^{-1}$ and corresponding loss tangent $\omega L/R$ measured at $T_b = 10$ mK and $\Phi = 0$ for the second (third) resonance in Sample 1 (2). The effective inductance (resistance) per single SNS junction is L/10 (R/10). Both frequencies are in the regime $\omega \approx E_T/\hbar$.

Sample	fn [GHz]	<i>R</i> [Ω]	<i>L</i> [nH]	2πf _n L/R
1	0.914	310 ± 30	3.4 ± 0.3	0.062
2	1.452	590 ± 50	3.1 ± 0.7	0.048

on gold-palladium SNS junctions. That is, they imply that an effective inductance of a few hundred picohenries per junction and a loss tangent of a few percent are observed around 1 GHz at millikelvin temperatures.

The accuracy of the admittance measurement is limited by uncertainty in the fit parameters f_n and Q_i , and the systematic device parameters measured in control samples (e.g. f_0 , C_i). The error bars of the *R* and *L* values throughout this article are propagated from the uncertainties in the fitted f_n , Q_i values at the 68% confidence level. There remains a 10% relative error in the reported *R* and *L* values from the systematic device parameter uncertainties at 1 GHz. This arises predominantly from chipto-chip variations in the unterminated resonance frequency, $\Delta f_0 \approx 3$ MHz, based on measurements of control samples. As frequency increases, Δf_0 has a larger effect on the uncertainty of *L* in Equation (2). For example, in Sample 2, the uncertainty $\Delta f_0 \approx 3$ MHz leads to a relative error in *L* of 70% at 10 GHz. For this reason, we concentrate on resonances near 1 GHz in the discussion below.

4. Theory

In the next section, we compare the experimental results to theoretical predictions^[18] based on the time-dependent Usadel equation.^[2] In the low-frequency and low-temperature regime $\hbar\omega$, $k_{\rm B}T \leq E_{\rm T}$ considered below, the imaginary part of the admittance of the junction is expected to be mostly determined by the adiabatic Josephson inductance associated with the supercurrent, i.e., the Φ derivative of the dc supercurrent. The real part, on the other hand, mainly arises from driven quasiparticle transitions in the junction. The availability of such transitions is sensitive to the density of quasiparticle states. In particular, the presence of a proximity-induced energy gap $E_{\rm g} \approx E_{\rm T}$ in the density of states should lead to an exponential increase in the resistance as $k_{\rm B}T$ decreases below $E_{\rm g}$.

However, the low-temperature values of *L* and R^{-1} we measure (Table 2) are dramatically larger than those predicted using the parameters considered in Reference [18]. This is evident from a cursory comparison of Figure 1 in Reference [18] to our $(\omega L/10)^{-1} \approx 6 R_N^{-1}$ and $(R/10)^{-1} \approx 0.3 R_N^{-1}$. The inductance per junction $L/10 \approx 300$ pH is also an order of magnitude higher than the expected adiabatic Josephson inductance $L_J = [2\partial_{\Phi}I_s(\Phi)]^{-1} \approx 50$ pH, where we approximate the dc supercurrent $I_s(\Phi)$ as $I_c \sin(\pi \Phi/\Phi_0)$ and the critical current I_c as the ideal value 6.7 E_T/eR_N for $\Delta/E_T \approx 13$.^[4] Moreover—in the results below—we observe a weak temperature dependence of $R(\Phi = 0)$ measured near 1 GHz, which is in stark contrast to the theoretically predicted exponential dependence.

The observed values of *L* and *R* imply that the proximityinduced superconductivity is significantly weaker than expected. We consider two distinct scattering mechanisms as potential explanations for this. First, we include dephasing due to inelastic scattering by choosing a phenomenological relaxation rate Γ .^[18] Second, we include a spin-flip scattering rate Γ_{sf} which could arise from dilute magnetic impurities in the weak link.^[31] Specifically, we include the spin-flip scattering as an additional self-energy $\bar{\sigma} = -\frac{i}{2}\hbar\Gamma_{sf}\hat{t}_{sg}\hat{t}_{sg}$ in the equations defined



in Reference [18]. Considering spin-orbit impurity scattering in a similar approximation^[32] would not affect the superconducting proximity effect within the model considered here. Although quantitative details differ, both of the scattering mechanisms generally lead to increased dissipation and increased inductance. Increased dissipation occurs mainly due to the suppression of E_{g} , while increased inductance occurs mainly due to the increase in L_1 .

Theoretical work on the microscopic origin of the scattering rates in disordered metals is reviewed in References [33] and [34]. Experiments have also been carried out with high-purity metal wires.^[35,36] However, we are not aware of measurements on the gold-palladium alloy used here, which prevents direct comparison to existing literature. Instead, our goal is to estimate the scattering rates required for a qualitative match to the experimental results. We find that in order to reproduce the experimentally observed *L* or *R*, the phenomenological rates Γ and $\Gamma_{\rm sf}$ must be large, i.e., comparable to $E_{\rm T}/\hbar$ and $k_{\rm B}T/\hbar$.

Inelastic scattering and spin-flip scattering are not the only possible explanations for observing proximity-induced superconductivity that is weaker than what is predicted by the ideal Usadel-equation-based theory. Although we do not attempt to exhaustively cover all candidates, we note that the SN contact resistance in our samples is much smaller ($R_{\rm B} < 1 \Omega$) than the normal-state resistance ($R_{\rm N} = 15 \Omega$). While the smallness of the ratio $R_{\rm B}/R_{\rm N}$ does not conclusively exclude explanations based on imperfect interfaces, it limits them significantly.^[5,37]

5. Temperature and Flux Dependence near 1 GHz

Below, we compare the predicted and observed dependences of Z_{chain} on the bath temperature and magnetic flux. We choose to analyze two low-*n* resonances near 1 GHz, mainly because the *L* values we extract for them suffer the least from the uncertainty in f_0 .

Figure 4 shows the measured flux dependence of *R* and *L* for the third (n = 3) resonance in Sample 2. The bath temperature is $T_{\rm b} = 195$ mK, which should be high enough for neglecting the hot-electron effect, i.e., for assuming that $T \approx T_{\rm b}$. As expected, we observe that *R* and *L* are periodic in flux, and that the inductance *L* and the loss tangent $\omega L/R$ are minimized (maximized) at integer (half-integer) values of Φ/Φ_0 .

Figure 4 also includes theoretical predictions for two different rates of inelastic scattering. The weaker of the two rates ($\Gamma = 2.3 \ k_B T_b/\hbar$) reproduces $R(\Phi = 0)$ well and gives a reasonable prediction for its flux-dependent oscillations. Furthermore, if we could only measure changes in *L*, we might conclude that the predicted flux modulation of *L* is in fair agreement with the experimental data for this moderate value of Γ . However, the absolute value of the prediction for $L(\Phi)$ is several times smaller than the observed value at nearly all flux values. This highlights the importance of measuring *L* and *R* without offsets if theories are to be rigorously tested. Note that we can improve the agreement between the predicted and measured $L(\Phi)$, especially around integer values of Φ/Φ_0 , by using a very strong inelastic scattering rate of $\Gamma = 8k_B T_b/\hbar$ in the theoretical calculation. However, this value of Γ leads to a



Figure 4. Flux-dependent a) *L* and b) *R* measured near 1.4 GHz of the SQUID chain in Sample 2 at 195 mK. Dashed lines are theoretical calculations that include only inelastic scattering corresponding to a very high scattering rate $\Gamma = 8k_BT_b/\hbar$ (black dashed line) and a moderately high scattering rate $\Gamma = 2.3k_BT_b/\hbar$ (gray dashed line). Solid lines are calculations that include very strong elastic spin-flip scattering, $\Gamma_{sf} = 15E_T/\hbar$ (black solid line), and moderately strong scattering, $\Gamma_{sf} = 4.5E_T/\hbar$ (gray solid line). Both solid lines include an additional low inelastic rate of $\Gamma = 0.05k_BT_b/\hbar$. Note that the theory calculations represented by the black dashed line and black solid line are nearly indistinguishable in panels (a) and (b).

clear disagreement in the amplitude of the oscillations in $R(\Phi)$ as shown in Figure 4b.

Figure 4 also shows the theoretical predictions that include strong spin-flip scattering. By choosing Γ_{sf} appropriately, the predictions become nearly identical to the case of strong inelastic scattering. Therefore, the conclusions of the previous paragraph also apply to predictions where scattering is spin-flip dominated. Furthermore, the similarity of the predictions shows that, in this parameter regime, the source of additional dephasing is unimportant.

To gain further insight, we study the temperature dependence of $R(\Phi = 0)$ and $L(\Phi = 0)$ for one resonance from each sample near 1 GHz (see Figure 5). In addition to the measured data points, Figure 5 shows theoretical predictions with scattering parameters that-at 195 mK-are identical to those in Figure 4. However, we note that considerable freedom remains in choosing the temperature dependence of the scattering rates. Rigorously justifying a particular temperature scaling would require knowledge of the specific microscopic mechanism responsible for the scattering. However, as the theoretical predictions already disagree with the measured results at the phenomenological level at a fixed temperature (Figure 4), identifying any specific microscopic mechanism seems implausible. As instructive examples, we choose $\Gamma \propto T$ and a constant Γ_{sf} in Figure 5. Unsurprisingly, none of the predictions simultaneously matches the observed temperature dependence of L and R. Nevertheless, the experimental data in Figure 5 may serve an important role in testing alternative theories in the future.



Figure 5. Temperature dependence of a) $L(\Phi = 0)$ and b) $R(\Phi = 0)$ for Sample 1 near 0.9 GHz (filled triangles) and for Sample 2 near 1.4 GHz (filled circles). The scattering rates for the theoretical predictions are given in Figure 4.

6. Conclusions

The main discrepancy between theory and experiment can be summarized as follows. The proximity effect at $T \leq E_T/k_B$ and $\omega \approx E_T/\hbar$ is weaker than what is predicted by theory based on the Usadel equation.^[18] This disagreement manifests itself experimentally as measured *R* values that fall below theoretical predictions and measured *L* values that exceed theoretical predictions. As potential candidates for such loss of coherence, we considered inelastic scattering and spin-flip scattering in the weak link. However, we did not find choices of Γ or Γ_{sf} that would provide simultaneous agreement in *L* and *R*, neither in terms of flux dependence at a fixed bath temperature, nor in terms of temperature-dependence at zero flux bias. Furthermore, the scattering rates required for a match in either *L* or *R* are larger than expected for, e.g., electron–electron scattering in disordered systems.^[34]

We note that the discrepancies shown here are not in direct contradiction with the previous experiments^[22,23] and that both the SNS junctions and the measurement scheme presented here are very different from these preceding studies. Firstly, the weak link material is different than in the previous experiments. We cannot rule out the possibility of effects specific to gold-palladium^[38] that reduce coherence in the weak link. Secondly, we measure both the reactive and dissipative components of the electrical admittance without arbitrary offsets. In contrast, only changes in the admittance have been reported previously. Thus, our experimental technique provides a more stringent test of the accuracy of the theory and reveals quantitative disagreements more easily.

In conclusion, we reported measurements of microwave frequency admittance for gold-palladium SNS junctions, together with a comparison to quasiclassical theory for diffusive SNS weak links. These discrepancies between measurement results

and theoretical predictions suggest that dephasing caused by inelastic scattering, or elastic spin-flip scattering, is probably not the correct mechanism for explaining why the proximityinduced superconductivity is weaker than expected in our gold-palladium SNS junctions. Further theoretical work is required for reaching simultaneous agreement for the magnitude, temperature dependence, and flux dependence of both the dissipative and reactive parts of the admittance. Mechanisms that may need to be taken into account include imperfect interfaces,^[5,37] electron-electron and fluctuation effects in low-dimensional superconducting structures,[39,40] and paramagnon interaction.^[41] Magnetic effects could be particularly important in SNS junctions that include palladium, which is paramagnetic in bulk and can even become ferromagnetic in nanoscale particles.^[42,43] In general, the relationship between microscopic materials properties and coherence at microwave frequencies in normal-metal Josephson junctions should be clarified, both experimentally and theoretically. A productive experimental approach may be to first investigate systems such as Nb/Cu weak links that, based on previous experiments,^[4,44] are expected to behave in an ideal fashion at dc.

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7. Experimental Section

Resonator fabrication: The substrates were 4" (0.5-mm-thick) highresistivity (>10⁴ Ω cm) Si wafers with 300 nm of thermal oxide. First, a niobium thin film (thickness 200 nm) was sputter-deposited on the entire wafer. Next, the coplanar waveguide (CPW) structures were defined with AZ5214E positive photoresist that was reflowed in air at 150 °C for 1 min to ensure a positive etch profile of the resulting Nb features. Then CPWs were etched with an rf-generated plasma under a constant flow of SF₆ (40 sccm)/O₂ (20 sccm) gases at constant power.^[45] The remaining resist was removed with solvents and an additional O₂-plasma cleaning step. The 4" wafer was then coated with a protective layer of resist and pre-diced with partial cuts along device pixel outlines on the back of the wafer.

Capacitor dielectric fabrication: The Al₂O₃ dielectric for the on-chip Nb-Al₂O₃-Al capacitors C_C, C_i, and C_{gnd} was formed by atomic layer deposition with 455 cycles in a H₂O/TMA process at 200 °C, resulting in a thickness of 42 nm. The thickness was verified in ellipsometry using an index of refraction $n_{Al_2O_3}$ =1.64. Measurements of reference Nb/Al₂O₃/ Al capacitors yielded a capacitance per unit area of 1.4 fF μ m⁻².

Nanostructure fabrication: The gold-palladium nanowires and aluminum superconducting leads were fabricated by electron beam lithography in two separate evaporation and liftoff steps. In the first step, gold and palladium pellets were evaporated from the same crucible with an electron beam heater. Afterward, unwanted AuPd was lifted off with organic solvents. Prior to the evaporation of the Al leads, samples were cleaned in situ with an Ar sputter gun. Finally, after liftoff of the Al film, individual resonator pixels were snapped along the pre-diced lines and packaged for measurement.

The chemical composition of the gold-palladium material was determined with energy-dispersive X-ray spectroscopy for incident electron beam energies 5 keV, 10 keV, and 20 keV (**Figure 6**). The average Au:Pd atomic ratio (weight ratio) was approximately 3:2 (3:1).

Cryogenic Measurements: Measurements were carried out in a commercial cryostat with a base temperature of 10 mK. The transmission coefficient was probed with a vector network analyzer. The device input line had >100 dB fixed attenuation. For all measurements, the output signal was amplified by a broadband low-noise cryogenic amplifier and by additional room temperature amplifiers. For some measurements (e.g. Sample 2, n = 3) two cryogenic isolators were placed on the base cooling stage between the low-noise cryogenic







Figure 6. Measured composition of a section of gold-palladium alloy evaporated together with Sample 2. The material stack from top to bottom is gold-palladium (30 nm), Al_2O_3 (42 nm), SiO_2 (300 nm), and Si (500 μ m). For clarity, we crop the strong signal for Si at high electron beam energies. It peaks at 2.8 keV⁻¹ (5.4 keV⁻¹) for 10 keV (20 keV).

amplifier and the sample. Each sample was placed in a custom-printed circuit board and sealed within a metal enclosure. The external flux coil consisted of a superconducting solenoid with 100 turns that was fixed outside the metal enclosure. One Φ_0 period in Figures 4 and 5 corresponds to a current change of $\Delta I_{mag}\approx 7$ mA through the coil. Magnetic shielding surrounded both the enclosure and the flux coil in the case of Sample 2.

The measurement power incident at the transmission line input was approximately –128 dBm for the data shown in Figures 4 and 5. This drove a current of roughly 5 nA through the SQUID chain at $\Phi = 0$ for n = 3 of Sample 2. This is far below the estimated critical current of the SQUID chain. Furthermore, experimentally we ensured that we measured the linear response by making sure that the measured S_{21} was not sensitive to factor-of-two changes in the measurement power.

Normalized transmission coefficient: We defined the normalized transmission coefficient $S_{21}(\Phi)$ as $S'_{21}(\Phi)/[S'_{21}(\Phi_{ref})/S_{21,fit}(\Phi_{ref})]$, where $S'_{21}(\Phi)$ is the raw transmission coefficient, including contributions from the cabling and other external circuitry and $S_{21,fit}(\Phi_{ref})$ is the prediction of Equation (1) at a reference flux value $\Phi_{\rm ref.}$ First, dividing $S'_{21}(\Phi)$ by $S'_{21}(\Phi_{\rm ref})$ removes all flux-independent features introduced by the external circuitry and unintentional reflections. The ratio $S'_{21}(\Phi)/S'_{21}(\Phi_{ref})$ only includes information about the response of the flux-varying SQUID chain since the external circuit leads to identical contributions in both scans. Second, we multiply $S'_{21}(\Phi)/S'_{21}(\Phi_{ref})$ by $S_{21, fit}(\Phi_{ref})$ in order to remove the systematic contribution of the reference in the final S_{21} result, which would otherwise appear at all values of Φ as a static vertically inverted mirror image $(1/S_{21, fit})$ of the reference resonance. We emphasize that we also determine $S_{21,\,\,{\rm fit}}(\Phi_{\rm ref})$ itself by fitting a ratio of two scans. That is, we fit $S'_{21}(\Phi_{ref})/S'_{21}(\Phi_{ref}+\Phi_0/2)$ to the quotient of two instances of Equation (1), each with a different set of fit parameters. These fit parameter determine the ${\sf S}_{\rm 21,\ fit}(\Phi_{\rm ref})$ used in the normalization process described above.

In practice, the scan used as the reference alternates between $\Phi_{ref}=\Phi_0/2$ and $\Phi_{ref}=0$, changing from one to the other whenever Φ crossed $\Phi_0(1/4+k/2)$, where $k\in\mathbb{Z}$. This kept the resonance in the reference far from the resonance frequency at the Φ value being analyzed. These changes in Φ_{ref} caused the apparent discontinuities in the background color in Figure 2.

As shown in Figure 2b,c the normalization procedure revealed clear Lorentzian-like lineshapes for S_{21} , as one would expect for a resonator of this type. Raw $S'_{21}(\Phi=0)$ and $S'_{21}(\Phi=0.3)$ data are shown in Figure S3 of the Supporting Information for reference. Disentangling the SQUID chain response from the rest of the circuit is also aided by the fact that the SQUID chain is the only circuit element that responds to flux periodically, with a period that is identical for all of the identified resonances. In general, the external circuit and background noise do

not have such a response as we confirmed in measurements of control samples with no SQUIDs.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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