Long-Range Coherence in a Mesoscopic Metal near a Superconducting Interface

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(Received 5 July 1995)

We identify the different contributions to quantum interference in a mesoscopic metallic loop in contact with two superconducting electrodes. At low temperature, a flux-modulated Josephson coupling is observed with strong damping over the thermal length L_T. At higher temperature, the magnetoresistance exhibits $h/2e$-periodic oscillations with $1/T$ power law decay. This flux-sensitive contribution arises from coherence of low-energy quasiparticle states over the phase-breaking length L_φ. Mesoscopic fluctuations contribute as a small h/e oscillation, resolved only in the purely normal state.

PACS numbers: 74.50.+r, 73.20.Fz, 73.50.Jt, 74.80.Fp

In a disordered metal at low temperature, electronic coherence persists over the phase-breaking length L_φ [1]. Weak localization, which consists in electron coherent backscattering along a closed diffusion path, induces corrections of the conductance of order the quantum of conductance e^2/h. The sensitivity of this process to an Aharonov-Bohm flux leads to $\phi_0 = h/2e$ periodic oscillations of the resistance of a mesoscopic loop [2,3]. Hybrid systems made of normal (N) and superconducting (S) materials are the scene for new physics, due to the Andreev reflection and the proximity effect. At low temperature ($k_B T \ll \Delta$), incident electrons have an energy much smaller than the gap Δ of S and are Andreev reflected at the N-S interface into a coherent hole. Spivak and Kmelnitskii investigated the effect of Andreev reflection on weak localization in a S-N-S geometry [4]. The N metal conductance was predicted to be sensitive to the phase difference between the two superconductors with a period of π, leading to a $h/4e$ flux periodicity in a loop. Petrashov *et al.* [5] and de Vegvar *et al.* [6] measured phase-sensitive transport in mesoscopic N-S metallic systems. The interpretation of Ref. [5] results in terms of weak localization is not consistent with the large amplitude of the effect [7]. In fact, the proximity effect in such mesoscopic systems can lead to a zero-resistance state with a well-defined Josephson current [8] if N-S interfaces have high transparency. In a two-dimensional electron gas, Dimoulas *et al.* also observed, beyond the Josephson coupling, large effects of quasiparticle interference on the resistance [9]. Recently, there has been considerable interest in coherent transport through mesoscopic N-S tunnel junctions [10,11]. Confinement of electrons and holes by disorder in N induces coherent multiple Andreev reflections, which enhance the low-temperature subgap conductance [12]. This is exemplified by the flux modulation of the subgap current in the case of a fork-shaped S electrode [13]. Volkov showed that this behavior may be explained by the appearance, despite the barrier, of a small pair amplitude in N [14]. This suggests that the proximity effect could explain most of the surprising data on resistive transport in mesoscopic N-S devices, even if classical estimates fail to agree with experimental results.

At present, a clear identification of the different contributions to coherent transport in N-S systems is missing. In this Letter, we describe new experimental results revealing unambiguously the nature of the different contributions to the phase-sensitive current in a mesoscopic N metal with S electrodes. Our experimental situation is greatly simplified compared to others, since we consider only the conductivity of the normal metal, the role of the interface being restricted to providing Andreev reflection. We used a ring geometry similar to that of Ref. [5], in order to control the phase by an external magnetic field, and made a study as a function of temperature. We show under which conditions the Josephson coupling, weak localization, and other contributions prevail. One important result is in the intermediate temperature regime, when the thermal length L_T is much smaller than L_φ and the sample size. We find a phase-sensitive contribution with an amplitude described by a $1/T$ power law. This contribution, much larger than the weak localization contribution, results from the persistence of electron-hole coherence far away from the N-S interface.

The inset of Fig. 1(a) shows a micrograph of a typical sample, made of a square Cu loop in contact with two Al electrodes. The Cu loop and Al electrodes are patterned by conventional lift-off e-beam lithography in two successive steps with repositioning accuracy better than 100 nm. *In situ* cleaning of the Cu surface by 500 eV Ar^+ ions prior to Al deposition ensures us of a transparent and reproducible interface. We performed transport measurements in a Mumetal-shielded dilution refrigerator down to 20 mK. Miniature low temperature high-frequency filters were integrated in the sample holder

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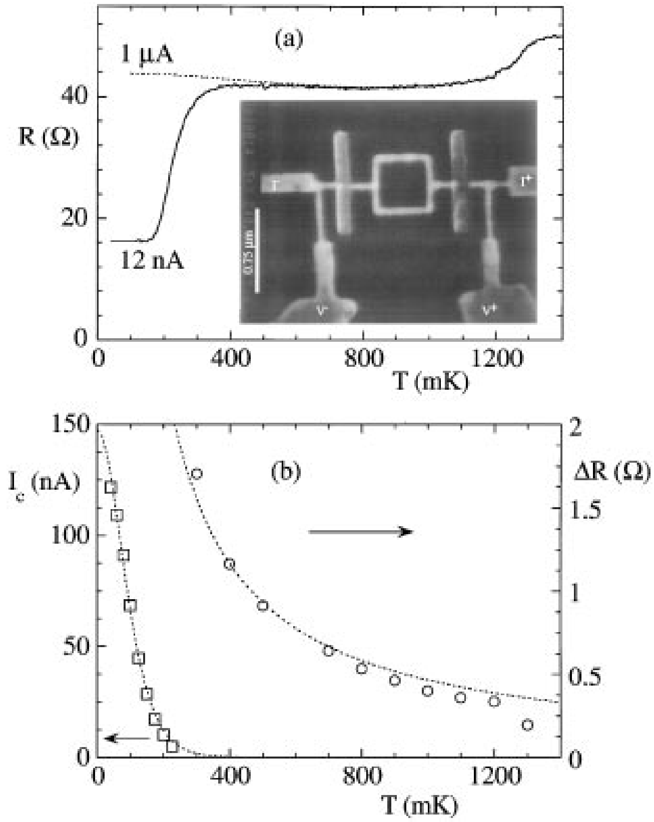

FIG. 1. (a) Temperature dependence of the resistance with a measurement current $I_{\mathrm{meas}} = 12$ nA and 1 μA. Inset: Micrograph of a typical sample made of a Cu square loop with four-wire measurement contacts, in contact with two Al islands (vertical). Diameter of the loop is 500 nm, width 50 nm, thickness 25 nm. Center-to-center distance between the 150 nm wide Al islands is 1 μm. The length L of the N part of the S-N-S junction is 1.35 μm. (b) Left scale: Temperature dependence of the critical current derived from Fig. 2 data with a 25 Ω differential resistance criteria. Dashed line is a guide to the eye. Right scale: Temperature dependence of the amplitude of the magnetoresistance oscillations, $I_{\mathrm{meas}} = 60$ nA. Dashed line is a $1/T$ fit.

[15]. We focus here on the experimental results of one sample representative of others. The 51 Ω normal-state resistance gives an elastic mean free path l_p of 16 nm and a diffusion constant D of 81 cm^2/s. The amplitude of the h/e oscillation in the normal state (see below) provides $L_\varphi = 1.9$ μm, so that the whole structure is coherent. The much smaller decay length of the pair amplitude in N is $L_T = \sqrt{\hbar D/2\pi k_B T} = 99$ nm$/\sqrt{T(\mathrm{K})}$. Figure 1 summarizes our results. Below the superconducting transition of Al at $T_c \approx 1.4$ K, the resistance of the sample decreases [Fig. 1(a)] by an amount corresponding to the coverage ratio of the Cu wire by Al islands ($\approx$20%). This behavior has already been met in previous experiments [8] and takes place provided the N-S interface resistance is low. This gives us a lower bound of 7% for the interface transparency t_0 and an upper bound of 230 nm for the barrier-equivalent length $L_t = l_p/t_0$ [16]. At very low temperature ($T <$ 250 mK), the sample resistance drops to a constant value of 16 Ω. The suppression of this drop by a large bias current suggests a Josephson coupling between the two S electrodes. The fraction of residual resistance (31%) can be related to the resistance of the normal metal between each voltage contact and the neighboring S island (23%), with an extra contribution due to the current conversion. Anticipating the discussion of the results, we show in Fig. 1(b) the temperature dependence of the two main contributions to transport. The Josephson current vanishes rapidly above 250 mK, revealing the exponential decay over L_T. The amplitude of the observed $h/2e$ magnetoresistance oscillations is plotted on the same graph. It represents more than 1% of the loop resistance and can be reasonably fitted by a $1/T$ power law. Let us now discuss these observations in more detail.

Figure 2 shows the current–differential-resistance characteristics for different temperatures between 42 and 225 mK. A sharply peaked feature indicates the switching of the loop into a resistive state. An unexplained additional structure can be seen at higher current. At the highest temperatures, a thermal rounding of the characteristic is visible. The shape of these curves can be qualitatively accounted for in a resistively shunted junction (RSJ) model with thermal fluctuations [17]. Solving the linearized Ginzburg-Landau equations, it is straightforward to calculate the pair current J_s between the two φ-dephased S electrodes. If the length L of N metal between the two S electrodes is large compared to L_T, we

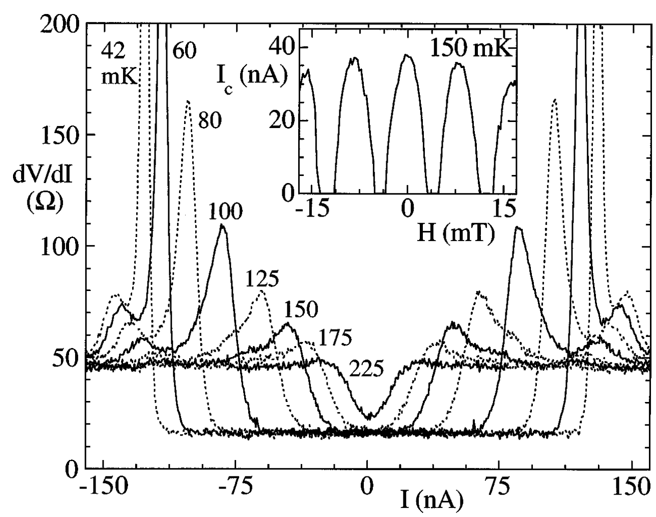

FIG. 2. Current–differential resistance characteristic at temperatures $T = 42$, 60, 80, 100, 125, 150, 175, and 225 mK, $I_{\mathrm{meas}} = 3$ nA. Inset: Magnetic field dependence of the critical current at $T = 150$ mK of the low temperature, low current $R = 16$ Ω resistance plateau. Differential resistance criteria are 35 Ω and $I_{\mathrm{meas}} = 3$ nA. The 8.25 mT periodicity of the oscillations corresponds to a quantum of flux $\phi_0 = h/2e$ in the 0.25 μm^2 loop area.

find a sinusoidal current-phase relation [18]:

$$J_s = J_0 \exp[-L/L_T] \cos(\pi\phi/\phi_0) \sin\varphi . \qquad (1)$$

where J_0 is a constant. The modulation of the maximum pair current by the magnetic flux ϕ with a period ϕ_0 is reminiscent of a superconducting quantum interference device (SQUID), although our mesoscopic geometry differs strongly from the classical design. Figure 2 inset shows the magnetic field dependence of the critical current at 150 mK. The 8.25 mT periodicity gives a $h/2e$ flux periodicity in the 0.25 μm^2 loop area, in agreement with (1). The observation of a Josephson coupling not found in Ref. [5] is attributed to the high transparency of our N-S interfaces.

In the high temperature regime (500 mK $< T < T_c$) the pair current is thermally suppressed. Calculating the thermal fluctuations of the pair current in the RSJ model [19], one expects exponentially small magnetoresistance oscillations:

$$\Delta R/R_N = -(1/8)(\hbar J_s/ek_BT)^2, \qquad (2)$$

which extrapolates to 10^{-8} at 1 K. In contrast, the magnetoresistance measured at various temperatures in this regime [see Fig. 3(a)] shows pronounced oscillations of $h/2e$ periodicity, which is consistent with Ref. [9]. With a sample resistance between the two S electrodes $R_N = 29\ \Omega$, the relative amplitude reaches about 1.4% at 1 K. No structure of half periodicity was met when measuring current amplitude was changed [20]. The amplitude of the low-field oscillations plotted in Fig. 1(b) shows a good agreement with a plain $1/T$ fitting law. The slight deviation of the data from the $1/T$ fit near T_c should be related to the depletion of the gap in S. This power-law dependence is a new result, in clear contrast with the exponential damping over L_T of the Josephson current. The characteristic features of the observed magnetoresistance oscillations are the following: (i) precise $h/2e$ periodicity with a resistance minimum at zero field, (ii) survival beyond the cutoff L_T of the Josephson effect, (iii) vanishing when the superconductivity of Al is destroyed above T_c or above critical magnetic field, (iv) same effect observed in samples with only one S island [17], and (v) a clear difference from mesoscopic fluctuations (period h/e) and weak localization $(h/2e)$ effects, both of amplitude e^2/h. For comparison, Fig. 3(b) shows the high-field magnetoresistance of the same sample when Al superconductivity is destroyed. We observe conductance fluctuations, with a h/e periodicity and a much smaller amplitude of $0.34e^2/h$, which gives $L_\varphi \approx 1.9\ \mu$m.

Our observations appear to be consistent with recent work from Zhou, Spivak, and Zyuzin [16] and previous work by Zaitsev [21] and Volkov and co-workers [14,22]. Zhou, Spivak, and Zyuzin showed that, beyond the expected strong suppression of the electric field in N over a length L_T from the N-S interface, corrections to resistive transport survive over distances up to $L_\varphi > L_T$. These corrections follow a power-law temperature dependence

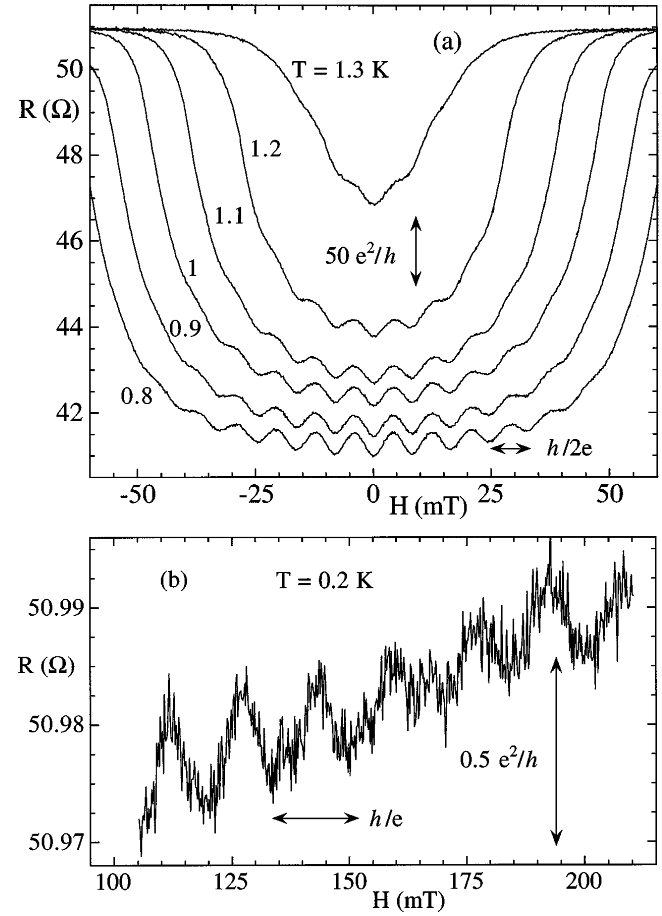

FIG. 3. (a); Low-field magnetoresistance for $T = 0.7, 0.8, 0.9, 1, 1.1, 1.2,$ and 1.3 K with $I_{\rm meas} = 60$ nA. $T = 0.7$ and 0.8 K curves have been shifted down by 1 and 0.5 Ω, respectively, for clarity. Oscillations of periodicity $h/2e$ and amplitude $16.7e^2/h$ at $T = 0.8$ K are visible. (b); High-field, and consequently normal-state, magnetoresistance of the same sample at $T = 0.2$ K, $I_{\rm meas} = 600$ nA. Conductance fluctuations appear, with a main component of periodicity h/e and magnitude of order $0.34e^2/h$, which gives $L_\varphi \approx 1.9\ \mu$m.

due to the long-range coherence of low-energy electron-hole pairs. Indeed, the decay length $L_\varepsilon = \sqrt{\hbar D/\varepsilon}$ for the pair-amplitude wave function $F(\varepsilon, x)$ diverges near the Fermi level $\varepsilon = 0$ [16]. Let us propose a simple picture for this effect. At the N-S interface, an incident electron is reflected into a hole of the same energy ε, but with a change in wave vector $\partial k = \varepsilon/\hbar v_F$ due to the branch crossing. After diffusion to a distance L from the interface, this induces a phase shift $\partial\varphi = \partial k v_F L^2/D = \varepsilon/\varepsilon_c$ between the electron and the hole. This means that at a distance $L > L_T$ electron-hole coherence is restricted to an energy window of width the Thouless energy $\varepsilon_c = \hbar D/L^2$, which is small compared to the width k_BT of the thermal distribution.

Resistive measurements in mesoscopic systems are strongly sensitive to the nature of electrical probes and the location of electron reservoirs, so that a quantitative description of the resistance behavior requires consideration of the particular geometry of the sample with its contacts. Nevertheless, the location of the N loop at a distance significantly larger than L_T from the N-S interface enables us to select the long-range component of the coherent quasiparticle states. The magnetic field creates an additional phase shift of 2π between the electron and the hole at flux $h/2e$, leading to $h/2e$-periodic modulation of the resistance. Here we do not observe an oscillation amplitude proportional to $L_T \propto 1/\sqrt{T}$, which would be the contribution of the whole N conductor between reservoirs [16], but a $1/T$ dependence related to the local contribution of the loop. This phase-sensitive contribution comes from the excess conductivity of those electron-hole pairs which have an energy close to ε_c. Both the temperature dependence and the amplitude of order 1% of the oscillations are indeed consistent with the ratio $\varepsilon_c/k_B T$. The similarity of this law with the expression of the local electric field at $L > L_T$ from Zhou *et al.* calls for further clarification. At very low temperatures ($T < \varepsilon_c/k_B$), one enters a new regime with the suppression of the conductance enhancement at low voltage [23]. This is not observed here due to the Josephson coupling, but is observed in samples with a single S island [17].

In conclusion, we have clearly identified the different components of the proximity effect in a mesoscopic metal near a superconducting interface. We demonstrated the crossover between the low-temperature Josephson coupling and a phase-sensitive conductance enhancement at high temperature. Compared to weak localization, this contribution has a larger amplitude and a distinct origin. The observed power-law dependence is consistent with the theoretical description of Zhou, Spivak, and Zyuzin [16] of long-range coherence of low-energy electron-hole pairs. Further theoretical progress is needed to have a clear physical understanding of the observed phenomena. This new effect could be very powerful in investigating phase-breaking effects in magnetic nanostructures [24] and electron-electron interaction in metals [16,23].

We thank P. Charlat for help in the experiment and J. Chaussy, A. Volkov, and A. Zaikin for stimulating discussions. We are indebted to F. Zhou and B. Spivak for clarifying discussions on their paper [16]. This work is supported by European Community under Contract No. CHRX-CT92-0068.

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