Experimental Observation of Bias-Dependent Nonlocal Andreev Reflection

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We investigate transport through hybrid structures consisting of two normal metal leads connected via tunnel barriers to one common superconducting electrode. We find clear evidence for the occurrence of nonlocal Andreev reflection and elastic cotunneling through a superconductor when the separation of the tunnel barrier is comparable to the superconducting coherence length. The probability of the two processes is energy dependent, with elastic cotunneling dominating at low energy and nonlocal Andreev reflection at higher energies. The energy scale of the crossover is found to be the Thouless energy of the superconductor, which indicates the phase coherence of the processes. Our results are relevant for the realization of recently proposed entangler devices.

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Andreev reflection (AR) is a well-known process that enables charge transfer across an interface between a normal metal and a superconductor [1]. At this interface, an incoming electron in the normal metal pairs with a second electron to enter the superconductor, resulting in a reflected hole. Past work has focused on the case of holes that are reflected back into the same electrode from which the incoming electrons originate. However, recent theoretical studies have considered the possibility that holes are reflected into a second, spatially separated electrode [2–11]. It was shown that this "nonlocal AR" process is equivalent to injecting two spin-entangled electrons forming the singlet state of a Cooper pair into two different normal leads [12]. In this way, nonlocal AR enables the realization of solid-state entanglers [13]—electronic devices capable of sourcing entangled pairs of electrons into nanoelectronic circuits—that are of interest for quantum information processing.

One way to investigate the occurrence of nonlocal AR relies on the following idea. Two normal metal electrodes are connected via two tunnel barriers (junctions $J1$ and $J2$) to one common superconducting electrode. If the separation of $J1$ and $J2$ is comparable to the superconducting coherence length ξ, an electron injected at energy $E < \Delta$ from the normal electrode of $J1$ can propagate as an evanescent wave through the superconductor and pair with an electron in the normal electrode of $J2$ [3]. This process results in a hole "reflected" into the second electrode, i.e., nonlocal AR. As holes have the opposite charge of electrons, holes undergoing nonlocal AR generate a voltage difference across $J2$ that has a sign opposite to that observed when the superconductor is in the normal state ($T > T_c^S$). Therefore, in principle, the detection of nonlocal AR is straightforward: $J1$ is used to inject current into the superconductor and $J2$ is used as a voltage probe to detect a voltage of the correct sign.

In practice, the situation is complicated by the occurrence of a second process competing with nonlocal AR: electrons injected from $J1$ can be transmitted into $J2$ without being converted into holes. This process is known as elastic cotunneling (EC) [4,11] and contributes to generate a voltage across $J2$ that has the same sign as that observed when the superconductor is in the normal state. Thus, the sign of the voltage measured across $J2$ depends on whether EC or nonlocal AR occurs with larger probability. The voltage measured at $J2$ may also vanish if cotunneling and nonlocal AR occur with exactly the same probability for all energies of the injected electrons. As some recent calculations predict [4,11] that this could in fact happen, it is not possible to anticipate which signal—if any—will be measured experimentally. For instance, in a recent experiment in which two ferromagnetic leads were used as normal electrodes, only the sign corresponding to EC has been observed [14].

In this Letter we report a clear experimental evidence for both nonlocal AR and elastic cotunneling using the experimental strategy just outlined. We find that the magnitude and the sign of the measured nonlocal voltage depend on the bias across the injecting junction. At low bias, the observed sign is the same as when the superconductor is in the normal state, indicating that EC dominates. At higher bias the sign of the voltage is reversed, which indicates the occurrence of nonlocal AR. The energy scale on which the sign reversal takes place corresponds to the Thouless energy of the superconducting layer. From this we conclude that the subgap microscopic processes of conduction, nonlocal AR and EC, are phase coherent.

A schematic representation of the devices used in our experiments is shown in Fig. 1(a). The structure is implemented in a Nb/Al multilayer sputtered on a thermally oxidized Si substrate using conventional Nb/Al technology [15]. The multilayer consists of two normal metal layers ($N1$ and $N2$, 50 nm Al layers) connected via two tunnel barriers to one common superconducor (S). Junction $J1$ is obtained by *in situ* oxidation of the $N1$ layer and subsequent deposition of Nb. Next, a thin (~5 nm) Al layer is sputtered on top of the Nb and oxidized *in situ*. Finally the top Al layer ($N2$) is deposited to form junction $J2$.

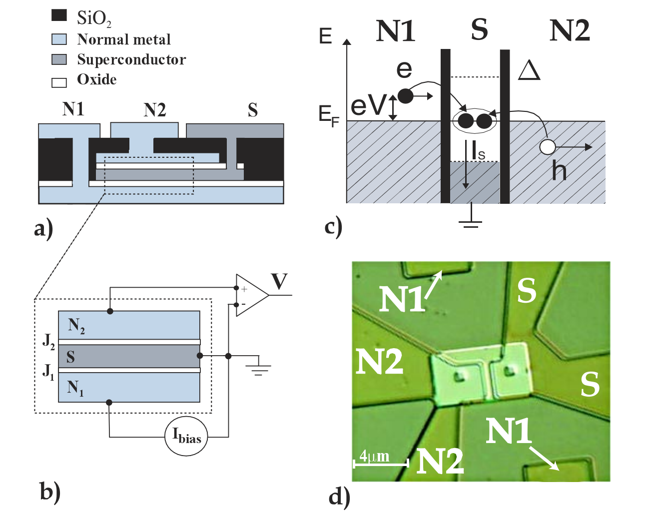

FIG. 1 (color online). (a) Schematic cross section of our sample (not to scale). Two normal electrodes ($N1$ and $N2$) are connected to a superconducting layer (S) via two tunnel barriers ($J1$ and $J2$), whose separation d is defined by the thickness of the superconducting layer. The concept of the measurement configuration is shown in (b): the current is injected through $J1$ and the nonlocal voltage is measured across $J2$. (c) illustrates the nonlocal AR process: an incoming electron from $N1$ is transmitted as a hole into $N2$ while a Cooper pair condenses in S. (d) Optical microscope image of one of our samples (top view). The rectangle in the center is where $J1$ and $J2$ are located; $N1$, $N2$, and S label the electrical contacts to the respective metallic layers.

The fabrication process used to pattern the multilayer relies on conventional photolithography combined with chlorine-based reactive ion etching. The junctions area is approximately $4 \times 8\ \mu\text{m}^2$. Independent electrical connections to the three layers are formed by deposition of a 200 nm thick Al/Nb layer on a SiO_2 mask followed by dry etching. We have checked the quality of the tunnel junctions by fitting the differential conductance with the usual BCS theory and found that the tunneling characteristics of junctions $J1$ and $J2$ do not show any substantial difference. This indicates that the superconducting properties of the Nb/Al layer (S) are uniform across its thickness.

In our devices the separation between the two tunnel barriers is determined by the thickness of the S layer, which can be controlled on the nanometer scale. This is crucial, since the separation of the tunnel barrier has to be comparable to the superconducting coherence length in S, $\xi \simeq \sqrt{\xi_0 l_e} = 10$–$15$ nm [16] (where $l_e = 3D/v_f \simeq 2$ nm is the elastic mean free path, the diffusion constant $D = 1.6\ \text{cm}^2/\text{s}$, and $\xi_0 = \hbar v_F/\pi\Delta$). An optical microscope image of one of our devices is shown in Fig. 1(d).

All the measurements were performed at $T = 1.6$ K or higher, with the aluminum electrodes $N1$ and $N2$ in the normal state ($T_c^{\text{Al}} \simeq 1.2$ K). In the experiment we send current through one of the junctions (e.g., $J1$) and measure the nonlocal voltage V^{nl} across the other junction ($J2$) while maintaining the superconductor at ground. The current bias has a dc component and an ac modulation with an amplitude of 1 μA at 19.3 Hz, and a lock-in technique is used to measure the ac component of the nonlocal signal. This corresponds to measuring the contribution given to the nonlocal voltage by only those electrons which have an energy $E = eV_{\text{dc}}$, where V_{dc} is the dc voltage across $J1$.

Figure 2 shows the $V_{\text{ac}}^{\text{nl}}$ measured as a function of V_{dc} at two different temperatures (above and below T_c), on a sample in which the superconducting layer is 15 nm thick (approximately equal to ξ). At $T = 22.5$ K, when the Nb is in the normal state, the sample can be simply thought of as a resistance network: the measured signal is large, because of the resistance of the thin Nb layer, and bias independent. Microscopically, the signal is due to electrons injected into the Nb that have a large probability to diffuse into the lead used as a voltage probe. At 1.6 K the Nb is superconducting and the Al in the leads is in the normal state. Now the nonlocal voltage is much smaller and it depends on V_{dc}. Specifically, $V_{\text{ac}}^{\text{nl}}$ reverses its sign at $V_{\text{dc}} = 270\ \mu$V and eventually vanishes at $V_{\text{dc}} \simeq 700\ \mu$V, thus on a bias range

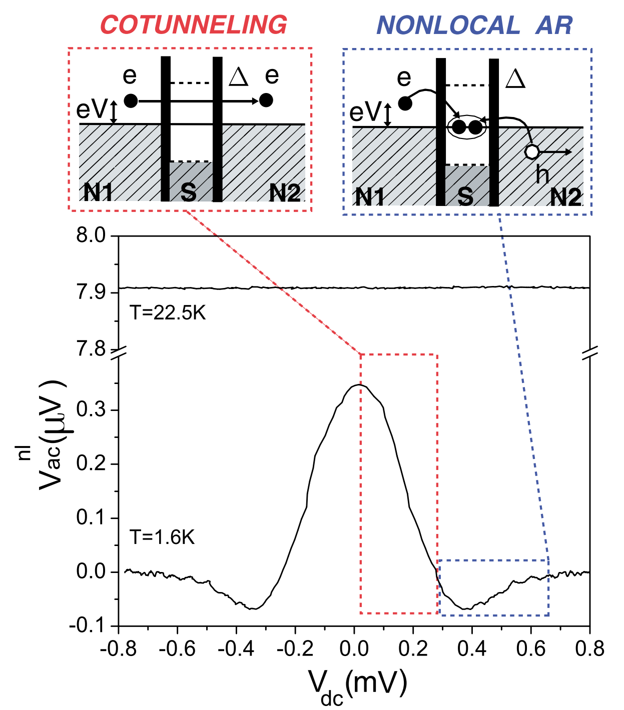

FIG. 2 (color online). The nonlocal voltage $V_{\text{ac}}^{\text{nl}}$ measured across $J2$, on a device with a $d = 15$ nm thickness of the superconducting layer, for two different temperatures. The upper curve is measured at $T = 22.5$ K—well above T_c^S—and shows a bias-independent nonlocal voltage due to electrons. At 16 K (below T_c^S), the nonlocal voltage is much smaller and depends on the bias V_{dc} across $J1$. At low bias, $V_{\text{ac}}^{\text{nl}}$ has the same sign measured in the normal state, indicating that elastic cotunneling dominates. At higher bias, the sign of $V_{\text{ac}}^{\text{nl}}$ is reversed, which indicates the occurrence of nonlocal AR.

much smaller than the superconducting gap [900 μV; see Fig. 3(b)].

To investigate if this signal originates from evanescent waves propagating below the superconducting gap, we have measured the nonlocal voltage in samples with different thickness d of the superconducting layer. Figure 3 compares the data measured in three samples where $d =$ 15, 50, and 200 nm, respectively. For the 50 nm sample, a nonlocal signal reversing sign with increasing dc bias is still visible at a bias range much smaller than the superconducting gap. However, the magnitude of the signal is approximately 20 times smaller than for the sample with $d = 15$ nm. For the sample with a 200 nm thick superconducting layer, no nonlocal signal is observed. These observations indicate that V_{ac}^{nl} is very rapidly suppressed with increasing the thickness of the superconductor, as expected for evanescent waves.

The comparison of different samples additionally shows that the energy scale on which the nonlocal signal reverses its sign (and eventually disappears) becomes smaller for a

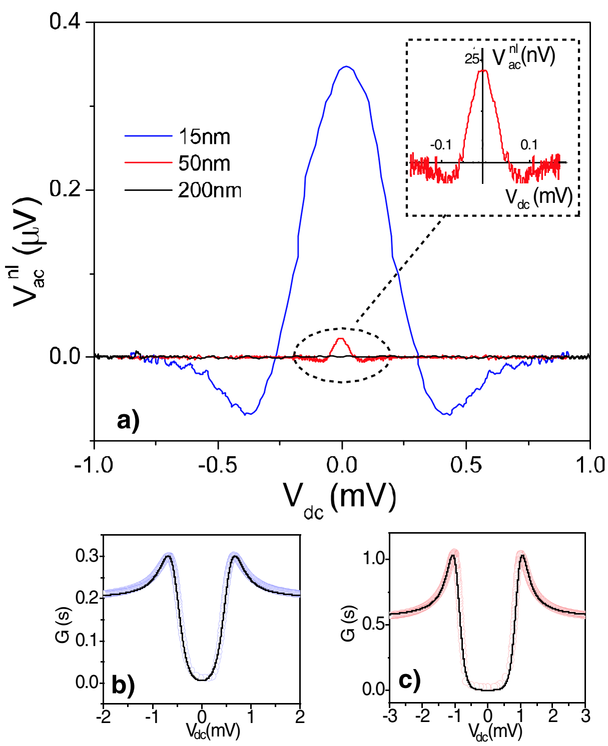

FIG. 3 (color online). (a) Nonlocal voltage V_{ac}^{nl} measured at $T = 1.6$ K on three samples with different thickness of the superconducting layer ($d = 15$, 50, 200 nm, with a normal state resistance of 4.8, 1.7, and 0.9 Ω respectively). Panels (b) and (c) show the tunneling characteristics of junctions, measured in two devices with $d = 15$ and 50 nm, respectively. The solid line is a fit based on the BCS density of states and shows that good agreement is found with $\Delta = 0.9$ and 1.45 mV for the two different thicknesses of the Nb layer [22]. The suppression of the gap in the $d = 15$ nm sample is typical of these thin superconducting films [16].

larger separation of the tunnel barriers. For the $d = 15$ nm sample the zero crossing energy is $\simeq 300$ μeV and for the $d = 50$ nm it is $\simeq 50$ μeV (see Fig. 3). These values correspond well to the Thouless energy $E_T = \hbar D/d^2$ of the superconducting layers, equal to $E_T \simeq 450$ μV and to $E_T \simeq 45$ μV for the $d = 15$ nm and the $d = 50$, respectively. The fact that the Thouless energy determines the behavior of V_{ac}^{nl} indicates that the signal originates from quantum-mechanically phase coherent processes. This is to be expected, since the transit time τ_{tr} of electrons injected from $J1$ and transmitted into $J2$—as electrons or holes—is $\tau_{tr} \simeq d^2/D \simeq 1$–10 ps, much smaller than the inelastic electron-phonon ($\tau_{ph} \simeq 1$ ns at 1 K in Nb) and electron-electron ($\tau_{ee} \simeq 0.1$ ns) interaction times [17].

Finding that E_T is the relevant energy scale in our measurements also gives an indication that nonequilibrium effects [18] in the superconductor do not play a relevant role in determining the behavior of V_{ac}^{nl}. In fact, these effects depend on the quasiparticle injection rate and relaxation times, whose energy dependence is not strongly influenced by phase coherent propagation in the superconductor (and thus by E_T). Note also that nonequilibrium effects normally become more relevant at higher bias voltage (when the amount of injected charge is larger), whereas the amplitude of the signal V_{ac}^{nl} is maximum at $V_{dc} = 0$ V and vanishes for V_{dc} well below the gap. The absence of nonequilibrium is consistent with the low transparency of our tunnel barriers ($T \approx 10^{-5}$) and with the fact that quasiparticles are injected with energies well below the superconducting gap. In contrast to quasiparticles occupying states above Δ, which may have very long relaxation times, quasiparticles with $E < \Delta$ decay very rapidly on the scale of h/Δ.

Having established the absence of significant nonequilibrium effects, we conclude that the measured nonlocal voltage V_{ac}^{nl} is due to phase coherent elastic cotunneling and nonlocal AR. EC is predominant at low bias whereas nonlocal AR dominates at higher bias, where the sign of V_{ac}^{nl} is negative. That the effect is large and present in all samples (approximately 10 samples with $d = 15$ nm and 50 nm have been studied) demonstrates that the sign reversal in the nonlocal voltage is not just a sample-specific effect, as has been observed in InAs/Nb structures [19].

The measured temperature and magnetic field dependence of V_{ac}^{nl} (see Fig. 4) are consistent with this interpretation. V_{ac}^{nl} increases with lowering T similarly to what one would expect from the convolution of a thermally smeared Fermi distribution with an energy dependent transmission probability (excluding the possibility that the signal is due to quasiparticle propagating above the gap). The signal is suppressed by a magnetic field applied parallel to the superconducting layer at $B \simeq 0.5$ T, which is much smaller than the critical field of our S layer (higher than 6 T [20]). Since Δ is only slightly reduced (few percent) by such a field, we believe that the main effect of B is the breaking of

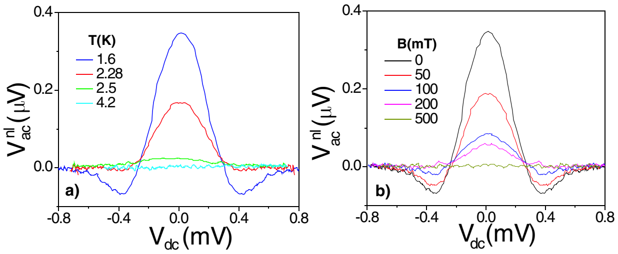

FIG. 4 (color online). (a) Temperature and (b) magnetic field dependence of the nonlocal voltage $V_{\mathrm{ac}}^{\mathrm{nl}}$ measured as a function of V_{dc}, on a sample with $d = 15$ nm.

time reversal symmetry for the electron-hole wave injected into the superconductor. Note, however, that at 0.5 T the magnetic flux enclosed by typical electron-hole trajectories in the superconductor (d is smaller than the magnetic penetration length in Nb for all samples) is only approximately $0.2 \times \phi_0$.

Our observation of a nonlocal signal shows that the cancellation of the contribution to $V_{\mathrm{ac}}^{\mathrm{nl}}$ due to nonlocal AR and EC does not occur in the samples investigated here. This cancellation was theoretically found in models that neglect the effect of Coulomb interaction [4], whereas calculations made for different systems in which interactions in the leads play a relevant role [5,7,9] did all predict the occurrence of visible effects. Since the effect of Coulomb interaction on electronic transport is visible in large-area tunnel junctions of size comparable to ours [21], we believe that Coulomb interaction may also be relevant here. A quantitative interpretation of our experimental results will require the analysis of theoretical models more sophisticated than those considered until now, which may have to address aspects of our samples that have not been considered so far (e.g., a gradient in the phase of the superconducting order parameter or a small subgap density of states induced by the presence of the normal electrodes).

In conclusion, we have reported clear experimental evidence for the occurrence of nonlocal Andreev reflection and elastic cotunneling through a superconducting layer. Our results show that these processes are phase coherent and strongly depend on the energy of the injected electrons. These findings are relevant for recent theoretical proposals of quantum entangler devices that aim at injecting into two spatially separated normal metal leads the spin-entangled electrons forming a Cooper pair. In this context, the energy dependence of the probability for nonlocal Andreev reflection may provide a new way to control the output of these entanglers.

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