EUROPHYSICS LETTERS 15 November 2001

Europhys. Lett., **56** (4), pp. 590–595 (2001)

Inverse proximity effect in superconductors near ferromagnetic material

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(received 20 April 2001; accepted in final form 27 August 2001)

PACS. 74.50.+r – Proximity effects, weak links, tunneling phenomena, and Josephson effects.

Abstract. – We study the electronic density of states in a mesoscopic superconductor near a transparent interface with a ferromagnetic metal. In our tunnel spectroscopy experiment, a substantial density of states is observed at sub-gap energies close to a ferromagnet. We compare our data with detailed calculations based on the Usadel equation, where the effect of the ferromagnet is treated as an effective boundary condition. We achieve an excellent agreement with theory when non-ideal quality of the interface is taken into account.

Proximity effect (PE) is a phenomenon where the order parameter of a material in an ordered phase leaks to another material which has no such order. A number of experiments have investigated the PE in superconducting-normal (S-N) mesoscopic structures [1]. Here a finite pairing amplitude is formed in the N side, with the decay length $\xi_{\rm N} = \sqrt{\hbar D/2\pi k_{\rm B}T}$ set by the dephasing of correlations between electrons and Andreev-reflected holes having slightly different momenta. At typical experimental conditions at low temperatures, the effect can survive up to large distances of several hundred nanometers. The experiments in non-magnetic metals have been generally well understood within the framework of the quasiclassical theory of inhomogeneous superconductivity [2]. Another example of a proximity effect occurs in systems composed of ferromagnetic (F) and non-magnetic materials, where the spin polarization leaks from F [3], but its decay length is typically very small. Going further, one may also consider the combination of two differently ordered materials, such as superconductors and ferromagnets.

In this letter, we study the mutual proximity effects of superconducting and ferromagnetic materials by measuring the local density of states in the superconducting side of the SF interface formed by superconducting Al and ferromagnetic Ni. Since the ferromagnetic and singlet superconducting order parameters try to exclude each other, the direct proximity effect is expected to get suppressed [4] so that the pairing amplitude diffusing into a ferromagnet

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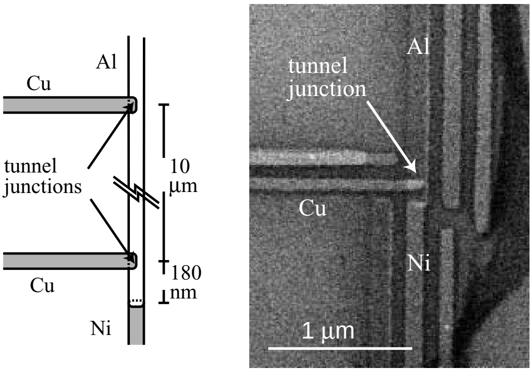

Fig. 1

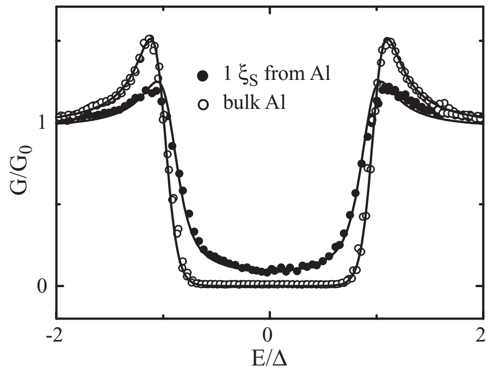

Fig. 2

Fig. 1 – Schematics of the sample, and SEM micrograph showing the region near the SF interface. The pattern was chosen so that the additional replicas due to shadow evaporation did not interfere with the desired pattern. The width of the lines was 100–150 nm.

Fig. 2 – Normalized differential conductance G/G_0 of the SIN junction, when properties of S are affected by the proximity of a ferromagnet. The circles are the experimental data at a distance of 180 nm (1.2 ξ_{S}) from the interface, and that for the bulk Al, shown for reference (measured at $T = 100$ mK). The data are normalized to the conductance value G_0 measured at 1 mV. The solid curves are the best fits to theory, with $\alpha = 7.5$ (implying $R_{\mathrm{env}} = 870\ \Omega$), $E_{\mathrm{C}}/\Delta = 1.5$, $r_b = 3$. The gap of Al was $\Delta = 0.22$ mV.

should decay over microscopic distances $\xi_{\mathrm{F}} = \sqrt{\hbar D/2\pi k_{\mathrm{B}} T_{\mathrm{Curie}}}$ due to the considerable difference in the momenta of spin-up and spin-down quasiparticles. A spectroscopy experiment in a low-T_{Curie} ferromagnet [5] agreed with the theoretical picture of this short-range effect. However, some recent experiments [6–9] appear to indicate the existence of a long-range (*i.e.* a penetration depth comparable to ξ_{N}) proximity effect into a ferromagnet and the situation has thus remained elusive. Here we report the first measurement on an inverse phenomenon, the modification of the BCS density of states in mesoscopic superconducting strips of Al under the influence of the proximity effect of a classical ferromagnet (Ni). This is rather a result from the expected suppression of the proximity effect: since the pairing amplitude vanishes at the SF interface, its value within a short distance from the interface is also much smaller than in the corresponding case of a finite proximity effect into a normal metal.

We use tunnel spectroscopy with tunnel probes at fixed positions to measure the differential conductance on the S side of the interface. At zero temperature, and for a vanishing charging energy of the tunnel contact, the differential conductance $G(E)$ with an N metal probe is proportional to the density of states (DOS). This way we can probe how the DOS in a superconductor is affected by the proximity effect. The DOS on the N side of an SN proximity system has been measured with fixed tunnel probes by Gueron *et al.* [10], who observed a dip in the N metal DOS up to one μm from the interface, with a quantitative agreement to theory. Several STM experiments capable of spatial mapping of the DOS have been carried out in short-coherence length layered superconductors [11–13], and recently in Nb-Au structure [14].

We used a sample structure shown in fig. 1. In the sample, we have two tunnel spectroscopy probes: one at a distance of approximately 180 nm from the transparent SF interface (called the interface junction), and one 10 μm from the interface (called the bulk junction). The latter tunnel junction is assumed to be located at a site with properties of bulk Al. The sample was fabricated on oxidized Si wafers by electron beam lithography and shadow evaporation through PMMA/MMA copolymer mask. We first evaporated 20 nm of Al. To make the clean SF interface, we evaporated a 25 nm layer of Ni at a different angle. The depositions of the first two layers were done in a single UHV cycle at pressures below $5 \cdot 10^{-9}$ mBar, avoiding any delay between the successive layers to make a transparent interface. The Al wires were then oxidized in 0.3 mBar of O_2 for 5 minutes in order to make the opaque barriers for tunnel spectroscopy. Lastly, the tunnel probes were made by depositing 40 nm of Cu at a third angle. The process yielded resistances of approximately 1.5 MΩ for both tunnel junctions.

For electric transport measurements, the sample was cooled down to approximately 100 mK in a plastic dilution refrigerator. Conductance measurements for the tunnel probes were performed through carefully filtered coaxial leads. To measure differential conductance, we used lock-in techniques with 20-30 μV ac excitation added to a constant dc voltage bias applied to the tunnel probe to be measured.

The experimental results are shown in fig. 2. For the bulk junction, we obtained a BCS-like differential conductance, although the BCS peaks were somewhat lower than what would be expected simply by thermal broadening. The magnitude of the peaks was accurately accounted for only when we took into account the Coulomb charging effects (see below). For the interface junction, the peaks were lower, with a clear sub-gap conductance.

In the normal state, superconductivity being suppressed by perpendicular magnetic field, we observed a conductance dip around small bias due to Coulomb charging effects. Both tunnel probes showed a dip of nearly 40% due to a resistive environment created by the relatively resistive nickel wires.

The diffusion constants for the wires or the interface resistance were not directly measurable in the sample. We used test samples, made in a similar process, to measure resistivities for thin wires of Al, Ni and Cu, with the results $\rho_{\mathrm{Al}} = 1.7\ \mu\Omega$cm, $\rho_{\mathrm{Cu}} = 2.0\ \mu\Omega$cm and $\rho_{\mathrm{Ni}} = 32\ \mu\Omega$cm.

The density of states near a SF interface can be calculated from the Usadel equation [15] for the quasiclassical Green's functions in the diffusive limit. These functions can be parametrized using the θ-parametrization [2]. In the absence of supercurrents the retarded Green's function is $\hat{G}^{\mathrm{R}} = \cosh(\theta)\hat{\tau}_3 + i\sinh(\theta)\hat{\tau}_2$, where $\hat{\tau}_i$ are the Pauli matrices in Nambu space. In the case of translational invariance in the transverse directions, the Usadel equation then takes the form

$$\hbar D \partial_x^2 \theta = -2iE\sinh(\theta(E,x)) + 2i\Delta(x)\cosh(\theta(E,x)). \qquad (1)$$

Here D is the diffusion constant and $\Delta(x)$ is the superconducting pair potential, calculated self-consistently from

$$\Delta(x) = \lambda N_0 2\pi k_{\mathrm{B}} T \sum_{\omega_n} \sinh\theta(\omega_n, x), \qquad (2)$$

where N_0 is the density of states in the normal state, and the sum goes over the discrete Matsubara frequencies $\omega_n = (2n+1)\pi k_{\mathrm{B}} T$ and is cut off at the Debye energy. The attractive interaction between the electrons is characterized by the coupling parameter λ, which is a non-zero constant for superconducting materials and zero otherwise. The parameter $\theta(\omega_n, x)$ is calculated from eq. (1), where the energy E is replaced by $i\omega_n$.

To account for a possible non-ideality of the interface between the two materials in the experiment, eqs. (1), (2) have to be complemented by a boundary condition recently derived by Nazarov [16], relating the Green's function in the left side of the interface ($\hat{G}_1 = \hat{G}(x = 0^-)$)

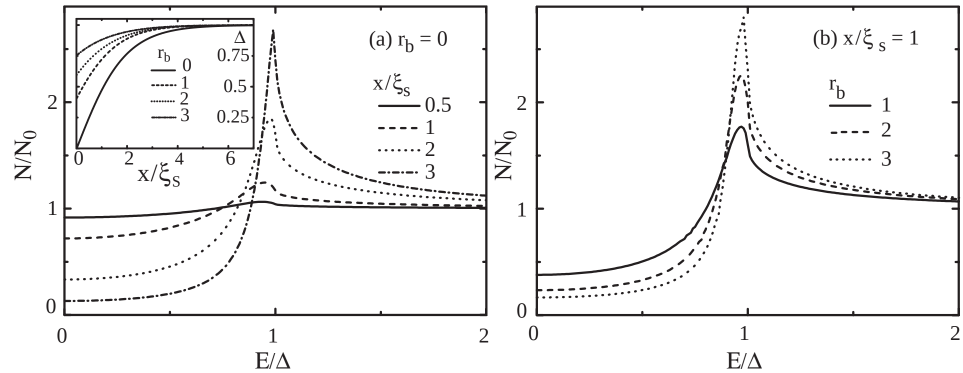

Fig. 3 – DOS of SF structures calculated on the S side, (a) with an ideal interface, $r_b = 0$, at distances $0.5\xi_S$–$3\xi_S$ from the interface; (b) at a fixed distance $x = \xi_S$ with varying interface parameters $r_b = 1$–3. In the calculations, the S order parameter was assumed not to penetrate F, *i.e.* the DOS in F was taken as constant N_0. The inset shows the self-consistent pair potential.

to the function in the right side ($\hat{G}_2 = \hat{G}(x = 0^+)$),

$$\sigma_1 \hat{G}_1 \partial_x \hat{G}(0^-) = \sigma_2 \hat{G}_2 \partial_x \hat{G}(0^+) = \frac{e^2}{h} \sum_n \frac{2\mathcal{T}_n \left[\hat{G}_2, \hat{G}_1\right]}{4 + \mathcal{T}_n \left(\left\{\hat{G}_2, \hat{G}_1\right\} - 2\right)}. \tag{3}$$

In a junction with many transmission channels, the distribution of the transmission eigenvalues $\mathcal{T}_n$ can be obtained from random matrix theory [17]. In the case of a dirty interface it is given by [18]

$$\rho(\mathcal{T}) = \frac{\hbar}{\rho_B e^2} \frac{1}{\mathcal{T}^{3/2}\sqrt{1-\mathcal{T}}}, \tag{4}$$

where ρ_B is the normal-state resistance of the interface per unit area. Integrating eq. (3) over the distribution of eigenvalues and over the cross-sections of the wires, we finally get the desired boundary condition

$$\partial_x \theta(0^+) = \frac{R_\xi}{R_{l\xi}} \partial_x \theta(0^-) = \frac{\sqrt{2}}{r_b} \frac{\sinh(\theta(0^+) - \theta(0^-))}{\sqrt{1 + \cosh(\theta(0^+) - \theta(0^-))}}, \tag{5}$$

where $r_b = R_B/R_\xi$ is the ratio between the normal-state resistance $R_B = \rho_B A_B$ of the interface with the cross-section A_B, and the normal-state resistance $R_\xi = \xi_S/\sigma_N A$ of a piece of the right-hand-side (S) wire with length ξ_S, conductivity σ_N and cross-section A. The corresponding resistance of the wire in the left (F) side of the interface is denoted by $R_{l\xi}$.

From the solution $\theta(E, x)$ to eqs. (1), (2), (5), we obtain the local density of states through

$$N(E, x) = N_0 \mathrm{Re}\{\cosh(\theta(E, x))\}. \tag{6}$$

In the ferromagnet, we assume the proximity effect to vanish within a few nm from the interface, and therefore set $\theta(x = 0^-) = 0$. In fig. 3 we plot the DOS at varying values of the interface parameter r_b and at varying distances to the interface. Since coupling to the interface decreases with increasing r_b, the DOS is very sensitive to the interface transparency.

To calculate the differential conductance of a tunnel probe, we have to take into account Coulomb charging effects due to the smallness of the tunnel junction capacitance. The charging effects depend on the impedance of the electromagnetic environment in terms of the function $P(E)$ which is the probability of exchange of energy between the tunneling electron and the environment. The forward tunneling rate through a single junction having the tunnel resistance R_T is given by [19]

$$\Gamma(V) = \frac{1}{e^2 R_\mathrm{T}} \int_{-\infty}^{+\infty} \mathrm{d}E\mathrm{d}E' \frac{N_0 N_\mathrm{S}(E'+eV)}{N_0^2} f(E)[1-f(E'+eV)]P(E-E'), \tag{7}$$

where N_0 is the DOS of the tunnel probe, taken as constant, and N_S is the proximity-affected DOS on the S side. The function $P(E)$ is calculated from an integral equation assuming a purely resistive environment [20].

For both tunnel probes, the parameters $\alpha = R_\mathrm{Q}/R_\mathrm{env}$ and the charging energy of tunnel junction capacitor E_C were determined as the values that gave the best fit to the normal-state conductance. Here, $R_\mathrm{Q} = h/4e^2 \simeq 6.5$ kΩ is the superconducting resistance quantum, and R_env is the resistance of the electromagnetic environment seen by the tunnel junction. Normal-state data of both junctions were fitted with the same values. These values were then used to fit Δ in bulk Al data, and we obtained a faultless fit with $\Delta \simeq 0.22$ mV and with the temperature $T = 100$ mK that was recorded in the experiment.

The distance between the interface and the first probe had some ambiguity of ± 50 nm because the tunnel probes had a finite width of 80 nm, and because the two metal films at the interface overlapped each other by approximately 50 nm. The average of the DOS over these distances was practically equal to the DOS in the middle of the tunnel probe, and we used the average distance (this distance is about $1.2\xi_\mathrm{S}$) in the analysis.

The normal-state and bulk-junction measurements yielded all the other important parameters of the system except the interface parameter r_b. For example, the electronic temperature in both bulk and interface junctions can be assumed the same due to the similarity of the probe resistances. Typical resistances reported for SF interfaces similar to our sample have been in the range 20 Ω–30 Ω [9]. For our sample made in UHV conditions, we expect $R_B \simeq 10$ Ω. Since $R_\xi \approx 2$ Ω, we would have $r_b \simeq 5$.

With a reasonable value of the interface parameter $r_b = 3$, we get an excellent fit with the theory, as seen in fig. 2. Note that this fit was obtained without taking into account such effects as the magnetic field due to the ferromagnet, penetration of the spin polarization in the superconducting side, or the emergence of a triplet proximity effect [21].

The effect of a stray magnetic field from the ferromagnet can be examined as follows. We expect the size of magnetic domains in the film to equal the thickness of the film, $d = 25$ nm. Since the dipolar field from a single domain decays with distance r as $(d/r)^3$, at relevant distances $r \simeq 200$ nm, the saturation field of Ni of 0.6 T has dropped down to 1 mT. Compared to the much larger critical field of the films (70 mT), this is unlikely to explain the enhancement of the DOS at low energies. To gain further confidence that the observed DOS was not affected by the stray magnetic field, we measured the conductance at various externally applied field strengths. At fields up to ~ 20 mT, the zero-bias conductance stayed practically constant while the width of the gap decreased. This behavior was similar in both the interface junction and the junction at bulk Al. Thus, stray fields even significantly larger than the estimated ones would not be likely to affect the zero-bias DOS, but would only reduce the gap. The case is strengthened by the fact that the gap was not reduced more than expected from the Usadel theory. These arguments also favor the idea that the domains were indeed of the estimated size.

In conclusion, we have measured the inverse proximity effect in a mesoscopic superconductor in contact with Ni, and compared the data with a self-consistent calculation based on the Usadel equation where we assume Ni to suppress the PE in a microscopic scale. We obtain a good agreement for the electronic density of states close to the interface only by taking into account the effect of the non-ideal interface.

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The authors would like to thank M. GIROUD and W. BELZIG for useful ideas and comments. This research was supported in part by the Emil Aaltonen foundation, and by the Human Capital and Mobility Program ULTI of the European Union.

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