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Local spectroscopy of a proximity superconductor at very low temperature

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Abstract. – We performed the local spectroscopy of a Normal-metal–Superconductor (N-S) junction with the help of a very low temperature (60 mK) Scanning Tunneling Microscope (STM). The spatial dependence of the local density of states was probed locally in the vicinity of the N-S interface. We observed spectra with a fully-developed gap in the regions where a thin normal metal layer caps the superconductor dot. Close to the S metal edge, a clear pseudo-gap shows up, which is characteristic of the superconducting proximity effect in the case of a long normal metal. The experimental results are compared to the predictions of the quasiclassical theory.

In the recent years, there have been a noticeable renewed interest in the proximity effect appearing at the junction between a Normal metal (N) and a Superconductor (S). In particular, the transport properties of the normal metal part were shown to exhibit a striking energy dependence with a long range persistence [1]. Another aspect of the proximity effect is the local modification of the energy spectrum. In the case of a N-S junction with a finite normal metal, the density of states in the normal metal is expected to exhibit a mini-gap : the density of states is zero within an energy window around the Fermi level [2,3]. The width of this mini-gap is smaller than the intrinsic superconducting gap of the superconductor. If two superconductors are connected to a short normal metal (S-N-S junction), the mini-gap will moreover depend on the phase difference between the two superconductors [4]. In the case of a normal metal with an infinite length, the density of states should exhibit a pseudo-gap [2,3]: the density of states is zero only at the exact Fermi level and goes approximately linearly with the energy close to the Fermi level. This behavior can be understood by arguing that some electron trajectories that travel close the interface never hit it, therefore do not couple to superconductivity and contribute to the density of states. Practically, the criteria of an infinite length for the normal metal should be understood as a length larger than the phase coherence length.

The local density of states in the vicinity of a N-S junction has been measured with the help of submicron planar tunnel junctions [5]. The appearance of a pseudo-gap in the N metal as well as the inverse proximity effect on the S metal side [6] were observed. A more local

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study is welcome in order to overcome the unavoidable spatial averaging of such conventional experiments. Indeed, the Scanning Tunneling Microscopy (STM) in the spectroscopy mode enables one to measure the very local density of states under the tip. Due to the technical complexity of (very-)low-temperature STM, the local study of mesoscopic superconductors is still in its infancy. Inoue and Takayanagi measured the tunneling spectra of a Nb-InAs-Nb system at 4.2 K [7]. Pioneering work by Tessmer et al. focused on the proximity effect in Au nano-sized islands on top of a NbSe₂ sample [8]. Levi et al. studied complex Ni-Cu-NbTi multifilamentary superconducting wires [9]. Recently Vinet et al. performed low-temperature spatially-resolved spectroscopy of Nb-Au structures patterned by lithography [10]. In this letter, we report on local spectroscopy measurements we performed on a proximity superconductor. We have been able to follow the evolution of the density of states from a fully-developed gap to a pseudo-gap as we move from the superconductor to the normal metal. Close to the N-S interface, a clear proximity effect is observed, both on the normal metal side and on the superconductor side of the interface. Eventually, we compare our experimental results to the predictions of the quasiclassical theory based on the Usadel equations.

Compared to previous works, we have been able to combine the very-low temperature (T < 1 K) conditions with the local probe technique. This guarantees a much improved energy resolution, and therefore the possibility to probe the proximity effect on a larger length scale, together with a high spatial resolution. Our very low temperature STM works at 60 mK in a dilution refrigerator [11]. It features both an atomic resolution and a large scanning range of $6 \times 6\mu m^2$ at low temperature. In the spectroscopy mode, this STM has shown an unprecedented energy resolution of 36 μ eV. This corresponds to an effective temperature of 210 mK which had to be introduced in the BCS fit of the spectroscopy data for plain Al and Nb layers. These fits were performed without any inelastic scattering parameter [12].

Our samples were made by successive in-situ evaporation of Nb (Superconducting below about 9 K) and Au or Cu (Normal metal). First, the Si sample substrate was introduced in the UHV evaporator with a patterned Si membrane clamped on it. This 5 μ m-thick Si membrane was previously patterned by e-beam lithography and deep Reactive Ion Etching. An array of circular holes with a periodicity of 4 μ m and a diameter of about 1.5 μ m were drilled throughout the membrane. During Nb evaporation, the Si membrane acted as a mechanical mask, so that only dots of Nb are deposited on the substrate. After deposition of 40 nm of Nb, the mask was removed in situ with the help of a UHV manipulator. Afterwards, an uniform layer of 20 nm of Cu or Au was deposited. The pressure was below 10^{-8} mbar during the few minutes between the two evaporations. This provides a highly transparent Nb-Au interface. Let us point out that no lithography or two-step deposition procedure was needed here, so that the Nb-Au interface is actually as clean as possible. We will concentrate here on one of the two Nb-Au samples we studied. Au was preferred in order to ensure a natural low oxidation of the sample surface during the transfer from the evaporator to the cryostat. One Nb-Cu sample showed a behavior similar to the Nb-Au samples. However, the more oxidized surface of Cu rendered the tunnel junction less stable and therefore more noisy.

Fig. 1b shows an STM image of the Nb-Au sample at 60 mK. Two Nb dots are clearly visible. The relief of the Nb dots is rather smooth. This is due to both the thickness of the mask and the residual distance between the substrate and the mask. This smooth relief is a drawback in terms of geometry simplicity but an advantage for STM imaging. We performed several series of spectroscopies as a function of distance from the center of a Nb dot by traveling along one line. During each series, the displacement speed was reduced to 10 nm/s and the scanning direction was kept fixed in order to reduce the piezo-electric hysteresis. Fig. 2a shows a representative selection of spectra taken during a single series. The surface profile of the same line is shown in Fig. 1b. Labeled arrows indicate the position of the spectra shown

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Fig. 1 – a : Profile of the sample surface extracted from image b) (line indicated indicated by the two arrows) together with a schematics of the sample geometry. Note that the vertical scale has been expanded by a factor of about 10 compared to the horizontal one. The locations where the spectra of Fig. 2a were measured are indicated, as well as the domains of application of the two geometry models used in the calculations (shown in Fig. 2b). In the "Bilayer" region, the sample is modeled by a series of (vertical) N-S bilayers, the thickness of S = Nb being given by the measured profile. In the "Proximity" region, the sample is modeled by a single (horizontal) N-S region. b : $2.6 \times 5 \,\mu m^2$ STM image at 60 mK captured with a 10 mV bias voltage and a 30 pA tunnel current. Two circular Nb dots are visible.

in Fig. 2. In the center of the Nb island (curve a), the density of states exhibits a clear gap, which is reminiscent of a BCS behavior. Compared to the bulk Nb gap value $\Delta_{Nb} \simeq 1.4$ meV, the measured gap is significantly reduced. This behavior is consistent with the measured critical temperature $T_c = 3$ K at the superconductivity onset. As we move away from the Nb dot center (curves b to d), the density of states first continues to exhibit a fully developed but reduced gap. This remains approximately true up to close to the curve e, which shows a clear pseudo-gap : the density of states goes approximately linearly to zero at the Fermi level. As the tip is moved further away (spectra e to j), the pseudo-gap width is reduced. We have been able to observe a pseudo-gap behavior in the density of states spectra over about 300 nm.

The proximity effect in diffusive metals can be described by the quasiclassical theory based on the Usadel equations [2–4,13,14]. In the usual θ parametrization, the complex angle $\theta(\epsilon, x)$ is related to the pair amplitude as : $F(\epsilon, x) = -i \sin \theta(\epsilon, x)$. The local density of states is expressed as :

$$n(r,\epsilon) = n_0 \Re e[\cos\theta(r,\epsilon)]. \tag{1}$$

The Usadel equations write :

$$\begin{cases} \frac{1}{2}\hbar D_S \delta_r^2 \theta + i\varepsilon \sin \theta + \Delta(r) \cos \theta = 0 & in S \\ \frac{1}{2}\hbar D_N \delta_r^2 \theta + i\varepsilon \sin \theta = 0 & in N \end{cases}$$
(2)

where D_N and D_S are the diffusion coefficients in N and S respectively. The inelastic and spin-flip rates were neglected. The gap $\Delta(r)$ in S is self consistently defined by :

$$\Delta(r) = n_0 V_{eff} \int_0^{\hbar w_D} tanh(\frac{\varepsilon}{2k_B T}) \Im m[\sin\theta] d\varepsilon, \qquad (3)$$

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Fig. 2 – a : Experimental spectra measured on locations a to j (see Fig. 1a) during a single scan along one line. The tunnel resistance was about 12 M Ω during the spectroscopies. b : Theoretical spectra calculated with the Usadel equations. The actual geometry was modelled as a series of N-S bilayers (curves a to d) and a single lateral N-S junction (curves e to j). An effective temperature of 210 mK was introduced. In the bilayer region, the measured thickness of the Nb layer ($d_S = 40, 21,$ 15 and 11 nm in a, b, c and d respectively) in units of $\delta_S = 27$ nm was introduced in the calculation together with the fixed Au layer thickness $d_N = 20$ nm = $0.3\delta_N$. In the proximity-effect region, the distance from the interface was a free fit parameter (see Fig. 3b).

where n_0 is the electron density, V_{eff} is the local interaction parameter and ω_D is the Deby frequency. In the case of a perfect transparency, the boundary conditions at the N-S interface include the continuity of θ at the interface and the spectral current conservation : $\sigma_S(\partial\theta/\partial x)_{x=0^-} = \sigma_N(\partial\theta/\partial x)_{x=0^+}$, where σ_S and σ_N are the conductivity in S and N respectively. In this work, we benefited from the numerical code developed by W. Belzig et al. [3], which solves the Usadel equation for a quasi-1D N-S junction and calculates the local density of states. The relevant theoretical parameters are : the gap Δ of the S metal, the mismatch parameter $\gamma = \frac{\sigma_N}{\sigma_S} \sqrt{\frac{D_S}{D_N}}$ and the thicknesses of the N and S layers in units of the characteristic lengths $\delta_N = \sqrt{\hbar D_N/2\Delta}$ and $\delta_S = \sqrt{\hbar D_S/2\Delta}$ respectively. Whereas the length δ_S is close to the superconducting coherence length ξ_S , the length δ_N does not stand for a coherence length of electron pairs in the normal metal. In order to analyze easily our experimental results, we modeled the different parts of our structures as quasi-1D junctions. On the Nb dot, we locally model the sample as an uniform bilayer of Nb and Au, see Figure 1b. Note that the vertical magnification by a factor of about 10 may make the reader underestimate the validity of this approach. In the remaining region labeled "Proximity", we model our sample as a single horizontal N-S junction, the interface position being a posteriori determined by the comparison with the theory.

Let us first consider the "Bilayer" region. The sample is locally modeled as a quasi-1D N-S junction with a constant length of N metal (the 20 nm thickness of the N layer) and a locally varying length of S metal (measured in the experiment). Since the Normal metal is small, a fully-developed gap is predicted, in agreement with the experiment. We assumed a

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Fig. 3 – a : Comparison between individual experimental data and two related calculated curves for an N-S junction with infinite N and S metals, using the parameters $\gamma = 1.1$ and $\Delta = 0.27$ meV. The position compared to the N-S interface is expressed in units of the characteristic length scales δ_N and δ_S . b : Comparison of the fit-derived distance from the interface with the actual (measured) position. Two data series taken on two different lines are shown and identified by squares and triangles symbols. The slopes of the adjusted lines give the length scales values $\delta_S = 50$ nm and $\delta_N = 94$ nm.

perfect interface transparency and no inelastic or spin-flip scattering. By fitting as a first step only the curve a, a good set of values was found to be $\gamma = 1.1$, $\Delta = 0.82$ meV, $d_S = 1.5\delta_S$ and $d_N = 0.3\delta_N$. The value of the γ parameter matches within the experimental accuracy the estimation of 1.2 based on the measured transport properties of the N and S layers. The magnitude of Δ is significantly smaller than the Nb bulk value, which we attribute to the small Nb thickness [15] and the special conditions of Nb evaporation. As the Nb and Au layers thicknesses are known, we can extract the length scales $\delta_S = 27$ nm and $\delta_N = 67$ nm. These values are in qualitative agreement with the respective expected values 41 nm and 55 nm. In this respect, let us note that the assumption of a diffusive motion over each layer thickness is only partially fulfilled since the mean free paths are of the order of the layer thicknesses. Curves for locations b to d in the "Bilayer" region were subsequently calculated by taking into account the fixed Au thickness $d_N = 20 \text{ nm} = 0.3\delta_N$ and the measured Nb layer thickness in units of $\delta_S = 27$ nm (inferred from curve a fit). As expected, the mini-gap amplitude is reduced as the thickness of the Nb layer decreases, the Au layer thickness being constant. The calculated curves are shown in Fig. 2b. In every curve, we introduced an effective temperature T = 210 mK in order to account for the experimental accuracy [11]. The agreement is very good, as both the mini-gap amplitude and the overall spectra shape are well described.

In the "Proximity" region, the measured spectra (curve e and beyond) show a filling of

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the density of states near the Fermi level and a peak shape that are not compatible with a bilayer model. This pseudo-gap behavior is the signature of the proximity effect in a N-S junction with a long normal metal N. In this "Proximity" region, we described the sample as a single infinite N-S junction extending laterally. We used the same γ parameter as in the "Bilayer" region. Again we consider a perfect interface transparency and zero inelastic and spin-flip scattering. In order to describe successfully the data, we had to assume a reduced gap value $\Delta' = 0.27$ meV. Compared to the value in the bilayer region, this reduced value may be understood as the effective gap of the Nb-Au dot treated as a whole. The experimental curves e to j were fitted within this model by considering the tip position as a free parameter. As an illustration, Fig. 3 shows the comparison between one experimental data and two theoretical curves which differ in the position compared to the interface. This position is expressed in units of the characteristic length scales $\delta_{N,S}$. The main fitting criteria was the density of states peak position, which actually moves towards the Fermi level as the position x referred from the interface is increased.

The validity of our description can be checked in Fig. 3b where we compare the actual position of the acquired spectra to the position extracted from the fit, in units of the relevant characteristic length. The data points follow a monotonous behavior, but with a significant scattering. The change of slope at the N-S interface reflects the difference in the characteristic length scales δ_N and δ_S . From the slope of the mean line we can draw through the data points on the N side (top part of Fig. 3), we can extract the value $\delta_N = 94$ nm. This corresponds exactly to the estimation based on the gap Δ' and the measured mean free path of 16 nm in Au. On the S side (bottom part of Fig. 3), the estimated length is $\delta_S = 50$ nm. Taking into account the reduced gap Δ' , it corresponds to a mean free path of 4.5 nm which is half the value estimated from transport properties of similar samples. In fact, it should be considered more as a property of the Nb-Au layer at its border than a property of the bare Nb film. This means that although our description seems both accurate and well-established in the N region, it is more difficult to relate the theoretical description on the S side to the real physical situation.

The location of the interface can be accessed from the fitting procedure. It appears that the interface is situated at an estimated Nb thickness of 6 nm. This means that the Nb layer is not superconducting when it is thinner than 6 nm. This behavior is expected in respect of the getter properties of Nb during evaporation. Let us also note that the density of states close to the interface is already much depressed compared to the "bulk" density of states in the superconductor. From the theory, this behavior is very sensitive to the mismatch parameter γ . This parameter is not intrinsic of the two materials since it not only depends on the N and S electronic densities ratio but also the mean free paths ratio. In Fig. 3, the scatter in the fitted position compared to the actual position can be envisioned as a signature of the extreme locality of the STM measurement. It is indeed visible from our whole set of data that the measured spectrum often changed abruptly as the surface is scanned. Some of these events are really local at the nanometer scale and show anomalies presumably related to point-like impurities. The other events feature step-like evolutions which are indeed in agreement with the overall behavior expected from the proximity effect. This behavior may be related to local scattering centers in the sample, like grain boundaries.

In conclusion, we have been able to probe the local density of states in the vicinity of a N-S junction at very low temperature. On the superconductor side of the junction, we observed a strong inverse proximity effect, with a density of states strongly modified compared to the BCS spectra. On the normal metal side, we observed a pseudo-gap of decreasing amplitude as the tip moves away from the N-S interface. The experimental data compares favorably with the spectra calculated from the Usadel equations, assuming a very simple geometrical model of

the complex sample geometry. In contrast with previous studies [5,10], we found that spin-flip and inelastic scattering could be neglected. This means that the related characteristic lengths should be larger than about 300 nm.

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