Nonequilibrium effects in superconducting necks of nanoscopic dimensions

H. Suderow\textsuperscript{1,2}, S. Vieira\textsuperscript{2}

\textsuperscript{1} Instituto de Ciencia de Materiales de Madrid, Consejo Superior de Investigaciones Científicas, 28049 Madrid

\textsuperscript{2} Laboratorio de Bajas Temperaturas, Departamento de Física de la Materia Condensada, Instituto de Ciencia de Materiales Nicolás Cabrera, Facultad de Ciencias, C-III, Universidad Autónoma de Madrid, 28049 Madrid-Spain

(November 14, 2018)

Abstract

We have fabricated superconducting connecting necks of Pb with a scanning tunneling microscope (STM) and studied their properties under magnetic fields near the transition to the resistive state. A striking phenomenology is found with two well defined conduction regimes as a function of the magnetic field. We discuss the possible origin of this behavior in terms of the interplay between the field dependence of the quasiparticle charge imbalance length $\Lambda_{Q*}$ and the length of the neck which is superconducting under field.

TO BE PUBLISHED IN PHYSICS LETTERS A
The study of very small superconducting systems is a topical research area showing many interesting aspects, relevant for possible applications in nanoelectronics [1]. Here we deal with connecting necks of nanoscopic dimensions between two superconducting electrodes. We expect that their properties are important to design of superconducting circuits at the smallest length scales, as they can help to understand the behavior of the connecting structures that are needed to operate those circuits. We build the necks by repeatedly indenting the tip of a scanning tunneling microscope into a sample (both being simple metals, like Pb), having the form of a constriction with a smallest cross section making the contact between both banks. These structures are too small to be fabricated using present nanolithographic techniques [2,4,5]. The mechanical, electronic transport, and superconducting properties have been studied in Refs. [2,4–8], where it was realized that the simultaneous measurement of the conductance and the displacement of the tip with respect to the sample give information about the form of the neck. Depending on the position on the surface where the repeated indentation is done, necks of different forms are obtained in-situ. For example, long structures, comparable to a short ”nanowire” attached to bulk electrodes [4]. The smallest cross section of the neck, the last contact, can be decreased towards the formation of a single atom point contact [8,9]. Transport is ballistic, as long as the mean free path is larger than the diameter of the contact, i.e. of the smallest cross section of the constriction [10].

One of the most interesting problems in small scale superconductors is the knowledge about their properties near the limits of the presence of superconductivity. For instance, the behavior of small wires near the critical temperature with an applied current has been a subject of intense research in the last decades, motivated both from the applied and fundamental point of views. As regards connecting necks, the transition at zero field turns out to be easily understandable using simple models [6]. Here we present results under magnetic fields, where we find a surprising phenomenology with anomalies in the differential resistance whose presence depends on the geometry. We propose that these anomalies are due to the fact that superconductivity is confined to nanoscopic length scales under magnetic fields. Indeed, in previous works we could show that in necks formed of Pb, which is a type I superconductor, above the bulk critical field \( H_c \), superconductivity is destroyed in the banks, but the neck remains superconducting when its lateral dimensions are smaller than the London penetration depth or the coherence length [7,8].

We use a STM within a \(^4\)He cryostat with a superconducting magnet and a conventional four wire I-V technique together with a Lock-In amplifier to measure the conductance. Considerable care has been taken to achieve a good resolution and to shield the apparatus from RF noise. The experimental procedure to obtain clean necks has been largely discussed [2,4,6–8], but we repeat here the main points. We cut a tip and a sample from a high purity (99.99\%) slab of Pb with a clean blade, immediately mount them on the STM and cool down in \(^4\)He exchange gas. Subsequently, we indent repeatedly the tip into the sample while recording the resistance \( R \) of the neck as a function of the elongation of the piezotube \( z \). \( R(z) \) is a staircase function whose overall form is different for each indentation and can be related to the topological form of the neck using the model of Ref. [3,4]. These authors found that one can model the whole constriction by a series of small slabs, each one of different radius and thickness, arranged symmetrically with respect to the center. We assume that the volume of the connecting neck remains constant. Then, the changes during the elongation of the neck are interpreted by a plastic deformation of the slab with the narrowest cross
section which breaks and forms a new slab with a smaller cross section. The smallest cross section \( A \) as a function of the elongation of the neck is easily calculated [4]. If the resistance is near the Sharvin limit \( R_S = \frac{h}{2e^2}(\frac{\lambda^2}{A}) \) with \( \lambda_F \) the Fermi wavelength) we can relate the measured \( R = R_S \) to \( A \) and each measured \( R_S(z) \) curve to a given \( A(z) \) function containing the form of the neck. In this work, we choose \( R \approx 5\Omega \), which corresponds to a contact with a diameter of about \( d = 6\text{nm} \). This is sufficiently small so that the conduction is near the ballistic regime (\( \ell > d \)) but still large enough so that the neck transits to the resistive state when a reasonably small current is applied (Fig.1). Note that in Ref. [8] we study the conduction properties of similar necks with three orders of magnitude smaller currents (the contact corresponding in that case to a single atom).

The Fig.1 shows two \( dV/dI(V) \) curves obtained at zero field corresponding to two different geometries. At zero voltage, a Josephson current flows and above the critical current \( I_{c,0} \) (limited by the contact), \( dV/dI \) shows anomalies located at \( V_n = \frac{2\Delta}{n} \) with \( n = 1, 2, 3, \ldots \) related to multiple Andreev reflection processes [11] (arrows in Fig. 1). The \( I(V) \) curve is shifted with respect to the resistive state ohmic I-V curve by the so called "excess current" \( I_{exc} \) (which is of the order of \( I_{c,0} \)), and smoothly disappears when the neck is heated by the strong current flow. Therefore, \( dV/dI \) increases slightly above \( 2\Delta \) and decreases at larger voltages, when the local temperature of the neck rises above \( T_c \) [6,12]. To estimate the local temperature, one can use the position in voltage of the \( n = 1 \) peak which is indeed at smaller voltages than expected from the position of the \( n = 2 \) peak (\( 2V_2 > V_1 \)) see arrows in Fig.1) [11]. Necks with larger opening angle (open points in Fig.1) are not heated in the current range that we study (\( V_n \) agrees in that case with the above expression, see also [8], we measure \( \Delta = 1.35\text{meV} \)).

Above \( H_c \) (Fig. 2), bulk superconductivity is destroyed, but the neck remains superconducting [8]. For \( V \leq 2\Delta \), the curves behave as the zero field curve, but with the features due to multiple Andreev reflections smoothed, in agreement with Ref. [8] where we discuss this in detail [13]. But the results for \( V > 2\Delta \) show a completely unexpected and very anomalous behavior.

High peaks appear in the \( dV/dI \) curve above \( 2H_c \) for \( 2\Delta < V < 6\Delta \)(Fig. 2). The peaks appear on top of the distorted \( dV/dI \) curve due to the above mentioned heating effects. A small voltage step ranging from \( 10\mu V \) to \( 100\mu V \), i.e. a small part (0.2 - 2%) of the whole voltage drop, is associated with each peak. The number of peaks varies between one and four. Each time we sweep \( V \) (from \(-15mV \) to \(+15mV \)) at a given magnetic field we do not find the peaks at the same voltage and their number is also different. The effect is independent on the direction of current flow. Note that in Fig.2 we only show the relevant voltage range and some characteristic curves for clarity. At larger magnetic fields (Fig. 2 c. and 2 d.), the number of observed jumps decreases to one and moves towards slightly smaller voltages. No peaks appear between \( 0.15 \) T and \( 0.2 \) T in this neck. If we build necks with larger opening angle \( \theta \), or stretch the neck towards smaller contacts, we also find a smooth behavior, without peaks. Therefore, the observed phenomenology cannot be associated to the (ballistic) conduction through the contact, but rather to the contribution of a part of the neck which transits to the resistive state under current flow. Indeed, in the necks with the smallest opening angle (around \( \theta \approx 30^\circ \), see Fig.1), it reasonable to expect that the conduction is quasiballistic, having a small contribution from regions of the neck outside the smallest contact. The peaks clearly indicate that the transition to the resistive state is
discontinuous, and this can be associated to the nucleation of phase slip centers within the neck.

The signature of phase slips are steps in the V-I characteristics. In phase slip wires, each step in the V-I curve gives a peak in $dV/dI$, and $dV/dI$ increases by a constant amount after every step \[14, 16\], that is proportional to the length $\Lambda_{Q*}$ where charge imbalance is found around the phase slip. $\Lambda_{Q*}$ is in general much larger than the coherence length and is given by \[17, 18\] $\Lambda_{Q*} = \sqrt{\frac{1}{3}v_F \ell \frac{1}{c} \tau_{E-P}}$ with $\ell$ being the mean free path, $v_F$ the Fermi velocity, $\tau_{E-P}$ the electron-phonon scattering time, and $\Delta$ the superconducting gap. In our case, we observe a small step in the $V-I$ curve and its associated peak in $dV/dI$, because the voltage drop associated to the transition to the resistive state of a part of the neck is in any case small. In order to obtain a more precise estimation, we would need to consider how the voltage drops on the whole neck, which is outside the scope of this paper. To gain further understanding, it is instructive to get an idea of $\Lambda_{Q*}$ in our situation. In order to explain the presence of several anomalies in our curves, we expect that $\Lambda_{Q*}$ should be of the order of or smaller than the maximal length of the necks (200nm). Nevertheless, simple estimates give a larger value for the lowest limit for $\Lambda_{Q*}$ in Pb $\Lambda_{Q*} > 430$ nm (we take $\tau_{E-P} \approx 2.5 \times 10^{-11}$ s \[15\] and $\ell \approx 10$ nm, see e.g. \[7\] for the complete T-H dependence of $\Lambda_{Q*}$). Note that this is in any case much smaller than $\Lambda_{Q*}$ in other phase slip wires (which can easily reach microns, see Refs. \[14, 15, 17, 19, 20\]) as $\tau_{E-P}$ is smaller in Pb than in other metals, but still larger than the length of our necks. Under field, $\Lambda_{Q*}$ is further reduced due to the pair breaking effect of the magnetic field which introduces a new time scale for relaxation so that $\tau_{E-P}$ is substituted by $\sqrt{\frac{\tau_{E-P}}{2(1/\tau_{PB}+1/\tau_{E-P})}}$ in the expression given above for $\Lambda_{Q*}$ ($\tau_{PB}$ is the pair breaking time; see \[17\]). In our case, the strong current flow should lead to an additional decrease of $\Lambda_{Q*}$ \[20\]. For instance, in the field range of Fig.2, a small pair breaking parameter (of $h/\tau_{PB} = 0.2T_c$), compatible with the reduction of the superconducting gap with field \[8\] already gives a reduction of $\Lambda_{Q*}$ by a factor of 2. So that $\Lambda_{Q*}$ can reach values comparable to the overall length of our structures. Although this gives only an order of magnitude estimate of the effect of the magnetic field, it becomes clear that the nucleation of a small number of phase slips within the neck is favored by the field. The irreproducibility in the number and position of the jumps is also compatible with the appearance of phase slips, as the situation is expected to be complex in a locally overheated environment.

At about 0.2 T (Fig.3) we observe another high peak which becomes most clearly visible at 0.25 T (Fig.3b) and then drops (Fig.3c). The corresponding anomaly in the V-I curve is again very small, of at most 100$\mu$V (at 2.5kG) and it has the same dependence on the geometry as the data shown in Fig.2, i.e. it is not present in necks with smaller contact or larger $\theta$ (see Fig.3c). We therefore also associate this peak with the transition to the resistive state of the neck. The difference with the peaks shown in Fig.2 is that, at a given field, when we sweep $V$ (from $-15mV$ to $+15mV$) we reproduce the behavior without changes nor hysteresis.

In this high field regime, we should take into account that the length $L_S$ of the superconducting part of the neck decreases significantly \[8\]. Indeed, we can separate the neck into small disks each one having a critical field given by $H_{critical} = 4\lambda H_{_c}$ \[7\]. Then, $L_S$ decreases with field as $L_S \sim (8\lambda H_c)/(H \tan \theta)$. According to \[17\], $\Lambda_{Q*}$ decreases more rapidly with field $\sim 1/\sqrt{H}$ in a simple, one dimensional system \[21\]. Therefore, as shown in the inset of Fig. 3c, at the highest fields, the length of the superconducting part of the neck can fall be-
low $\Lambda Q^*$. At the same time, charge imbalance created by the N-S interface leads to a greater voltage drop as compared to lower fields because it is created near the part of the neck with smaller radius. This could lead to the anomaly shown in Fig.3. The recent calculations of Ref. [8] on a similar neck confirm these estimations. It is shown that at 0.25$T_c$ (the same field where the anomaly is at its maximum), the N-S interface is well defined and located at a distance about three times the coherence length from the center of the neck. At larger fields, the height of the corresponding anomaly decreases as superconductivity is destroyed.

The whole phenomenology (Figs. 2 and 3) has been reproduced in different experiments, with the magnetic field parallel or perpendicular to the neck. It is robust and depends only on the geometry of the neck. We have measured several geometries (30° < $\theta$ < 70° and 5nm > $d$ > 0), and the phenomenology discussed above is only present in necks with a small opening angle and large contact diameter. No jumps in $dV/dI$ are observed in necks with smaller minimum radius, or necks with larger opening angle than in the case presented here. The temperature dependence of the curves shown in Fig. 1 is smooth: the bump disappears gradually without any sign of peaks in $dV/dI$ as expected from the decrease of the critical current with temperature.

Our interpretation gives a rough physical model for the observed behavior, and agrees only qualitatively with the results. But it presents a consistent model and a first approach to understand our system. We also note that hot spots due to heating [15] are excluded in our experiment as they should appear preferentially at zero field, where the bulk is superconducting and a much poorer heat conductor. We did not discuss effects associated with the presence of flux tubes as the flux going through our structure is in any case smaller than the flux quantum $\Phi_0$.

Recently, other anomalies that appear at zero field in N-S point contact junctions were explored [22]. However, the position in $V$ of these anomalies changes when decreasing the smallest contact radius, pointing towards a different origin. Some nanolithographated structures also present anomalies in the current-voltage characteristics whose origin is debated in Refs. [17,20,23,26]. For example, the resistance of some wires does not decrease at $T_c$ but grows above the normal phase value, and only drops at lower temperatures. The differential resistance shows peaks and anomalies in this regime. The form of these peaks and its dependence on defects or on the position and nature (normal or superconducting) of the nanofabricated voltage probes are studied in detail in Refs. [17,20,23,26]. The presence of phase slip centers and/or a N-S interface also seem to explain some of the observed behaviors. Also within this context, the recent theory of Ref. [27] explains a large critical field in wedge like structures, but it does not treat the transition to the resistive state.

Finally, it is also of interest to consider the possibility of quantum effects, because our necks are indeed potential candidates to observe quantum of phase slippage (their mean diameter and length is of the same order of magnitude as the nanowires discussed in Ref. [28]). Indeed, recently, the authors of Ref. [29] have measured the destruction of the superconducting state due to quantum phase slips in wires of similar diameter and somewhat larger lengths than the connecting necks presented here. They discussed the possibility of a dissipative phase transition (see [30]), depending on the resistance of the wires. In our case, however, we find anomalies that appear at high temperatures, indicating that the resistive centers are activated due to thermal rather than quantum fluctuations. Future measurements on larger necks and at lower temperatures might help to further study this point.
nevertheless, we expect that in necks, the presence of the normal (bulk) electrodes may renormalize the dimensionality of the system and decrease the importance of (quantum) fluctuations.

In conclusion, we have found a new phenomenology in the transition to the resistive state of superconducting connecting necks of nanoscopic dimensions under magnetic field. We have defined the conditions for the appearance of different conduction regimes. Nonequilibrium effects could explain the observed behavior.

Acknowledgments: We acknowledge discussions and help of F. Guinea, E. Bascones, A. Izquierdo, G. Rubio and N. Agraït. We also acknowledge financial support from the TMR program of the EC and the DGICyT under contracts ERBFMBICT972499 and PB97-0068.
REFERENCES

[1] Mesoscopic Electron Transport, Eds. Sohn, L.L., Kouwenhoven, L.P., Schön, G., 549-579. NATO ASI Ser. E, Vol. 345, Kluwer Academic, Dordecht (1997).

[2] N. Agraït et al. Phys. Rev. B, 47, 12345 (1993); N. Agraït et al. Phys. Rev. Lett., 74, 3995 (1995); G. Rubio et al. Phys. Rev. Lett. 76, 2032 (1996); N. Agraït et al. Thin Solid Films, 253, 199 (1994).

[3] We always check that the $R(z)$ curves have a continuous behavior. We stretch each neck until it breaks (tunneling regime), and check subsequently that the topological images of the surface in the conventional STM mode are the same for different tunneling resistances.

[4] N. Agraït et al. Phys. Rev. B, 48, 8499 (1993); C. Untiedt et al. Phys. Rev. B, 56, 2154 (1997).

[5] Brandbyge, M. et al., Phys. Rev. B, 52, 8499 (1995); Pascual, J.I. et al., Phys. Rev. Lett. 71, 1852 (1993); Landman U. et al., Science 248, 454 (1990).

[6] J.G. Rodrigo et al. Phys. Rev. B, 50, 12788 (1994).

[7] M. Poza et al. Phys. Rev. B, 58, 11173 (1998).

[8] H. Suderow et al., Europhysics Lett. 50, 749 (2000); H. Suderow et al., Physica C, 332, 327-332 (2000).

[9] E. Scheer et al. Nature 394, 154 (1998); Yanson et al., Nature 395, 783 (1998); Ohnishi et al., ibid, p. 781.

[10] A.V. Khotkevich, I.K. Yanson, Atlas of Point Contact Spectra of Electron-Phonon interactions in Metals. Kluwer Academic Publishers, Dordrecht (1995).

[11] M. Octavio et al., Phys. Rev. B, 27, 6739 (1983) is a pioneering work. See the References in [8] for more recent treatments.

[12] M. Tinkham et al. J. of Applied Physics, 48, 1311 (1977).

[13] Note that he overall length of the neck is limited by the elongation of the piezotube, 200 nm in our case. This is clearly several times $\xi$, estimated to be approximately 20nm, see [8].

[14] A.M. Kadin et al. J. of Low Temp. Phys., 33, 481 (1978).

[15] T.M. Klapwijk et al. J. of Low Temp. Phys., 27, 801 (1977).

[16] J. Skocpol et al., J. Low Temp. Phys., 16, 145 (1974).

[17] C. Strunk et al., Phys. Rev. B, 53, 11332 (1996); C. Strunk et al., Phys. Rev. B, 57, 10854 (1998).

[18] A. Schmid, G. Schön, J. of Low Temp. Phys., 20, 207 (1975); see also S.N. Artemenko et al., J. Low temp. Phys. 30, 487 (1978).

[19] J.M. Aponte, M. Tinkham, J. of Low Temp. Phys., 51, 183 (1983).

[20] C.J. Chien and V. Chandrasekhar, Phys. Rev. B 60, 3655 (1999).

[21] In a neck, we expect an even stronger variation of $\Lambda_{Q*}$ with field, due to the decrease of the length of the superconducting part, see [8].

[22] P.S. Westbrook and A. Javan, Phys. Rev. B, 59, 14606 (1999).

[23] Moshchalkov et al. Phys. Rev. B 49, 15412 (1994), Comment by I.L. Landau and L. Rinderer Phys. Rev. B, 56, 6348 (1997) and Reply ibid. p. 6352.

[24] Santhanam et al. Phys. Rev. Lett. 66, 2254 (1991).

[25] M. Park, et al. Phys. Rev. Lett., 75, 3740 (1995).

[26] K. Yu. Arutynov et al. Phys. Rev. B, 59, 6487 (1999).
[27] V.M. Fomin et al. Europhys. Lett. 42, 553 (1998), 46, 118 (1999).
[28] A.D. Zaikin et al. Phys. Rev. Lett. 78, 1552 (1997).
[29] A. Bezryadin et al., Nature, 404, 971 (2000).
[30] A. Schmid, Phys. Rev. B, 1506 (1983).
FIG. 1. The differential resistance $dV/dI(V)$ of a long (line) and a short (line and points) neck together with the corresponding I-V curves (inset). The geometry of the structures, as measured from the $R_S(z)$ curves, is sketched at the top of the figure. The arrows show the peaks due to the subharmonic gap structure [11]. The peak corresponding to $2\Delta$ appears in the long neck ($\theta_1 = 35^\circ$) at smaller voltages than in the short neck ($\theta_1 = 70^\circ$) due to self-heating, which leads to a decrease in $\Delta$. 
FIG. 2. $dV/dI(V)$ for different magnetic fields ($\theta = 35^\circ$, see line in Fig.1). The sketch on the right is to remark that above $H_c$ (0.05T at 4.2K) superconductivity is destroyed in the bulk, but not in the neck. This is discussed in detail in Refs. [7,8]. For $2H_c < H < 3H_c$ we observe high peaks in $dV/dI$ which appear at on top of the distortion of $dV/dI$ induced by heating.
FIG. 3. $dV/dI(V)$, normalized to the value in the resistive state as extrapolated from large voltages. In c.) we compare the neck showing the anomalous behavior (line in Fig.1) to a neck with the same $\theta$ but smaller $A$ (ten times smaller, $d = 2nm$, squares) and to a neck with two times larger $\theta$ (open points). None of these necks show an anomalous behavior. The inset in c.) sketches the expected decrease of $\Lambda_Q*$ and $L_S$ under field.