# Metallic to tunneling transition in Cu-Nb point contacts

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In a recent paper we proposed a detailed theory for the *I-V* curves of a normal-superconductor microconstriction, predicting a smooth crossover from metallic to tunnel-junction behavior. This paper describes experiments on Cu-Nb point contacts which span the crossover regime. We find excellent quantitative agreement between theory and experiment over a substantial range of contact resistances.

## I. INTRODUCTION

A high-quality tunnel junction results from a uniform, pinhole-free oxide layer separating two metallic electrodes. A high-quality microbridge results from a small, clean metallic neck connecting two bulk metallic "banks"; any oxide or other scattering mechanism in the bridge is to be avoided. Tunnel junctions and microbridges are both examples of weak links, meaning that on the average an electron has a small probability of transferring between electrodes. Although similar in this way, the different mechanisms of restricting electron flow produce distinctly different *I-V* curves. Still, one might imagine continuously evolving a tunnel junction into a microbridge by simultaneously reducing the area of the junction and the thickness of the oxide barrier in such a way as to hold the resistance roughly constant. Here we are interested in examining the basic physics of the crossover, but we recognize the possible practical applications of the theory. For example, oxide layers are rarely pinhole free, and microbridges are often "dirty." A thorough understanding of the deviations from the ideal limits, as well as of intentionally fabricated intermediate cases, may help guide efforts to improve devices using high-current-density small-area superconducting weak links.

We have previously published a theory[1,2] for the normal-superconductor (*N-S*) and the superconductor-superconductor (*S-S*) microconstriction. The purpose of this paper is to report on detailed experimental tests of the predictions of that model for the *N-S* case. Since we were interested in varying the strength of the interfacial barrier, we chose the point-contact technique as the simplest approach. Here, a sharpened wire (Nb) is pressed on a polished flat (Cu). The contact area is covered with a native, nonuniform oxide layer, and is rough on a microscopic scale. Thus, depending on the position along the surface, or the pressure of the point, the resistance can vary over 3—4 orders of magnitude, and the *I-V* curve can be transformed from metallic to tunneling type behavior. The next section provides a qualitative summary of our model; it is followed by a description of the experiment and the fit between the data and the theory.

## II. THEORY

Although a detailed theory for the *N-S* microconstriction is contained in Ref. 1, the qualitative behavior of the *I-V* curve can be easily understood with the simple physical arguments of this section. We may picture a three-dimensional microconstriction as a hole of radius $a$ in an insulating screen separating two bulk metals. If the mean free path $l$ satisfies $l \gg a$, then the resistance is due to the constriction impeding the ballistic transport of electrons through the hole. This is known as the Sharvin resistance[3] $R_0 = \rho l / 4a^2$. Since the $\rho l$ product is approximately temperature independent, the resistance depends only on the size of the hole. We refer to this situation as a "pure" metallic contact.

Now consider an *N-S* interface, where for simplicity we restrict attention to $T=0$, the $\rho l$ product is assumed to be the same in both metals, and there is no applied voltage. What will happen when an electron from the normal metal, with energy $E < \Delta$, tries to pass through the constriction and enter the superconductor? The electron cannot continue through as a quasiparticle, since the excitations in a superconductor have a minimum energy of $\Delta$. It also cannot be reflected as an electron, since this would imply the net current was zero, contrary to our expectation of an *N-S* interface as a good conductor. Instead, what occurs is Andreev reflection,[4] that is, the electron reflects as a hole in the normal metal while simultaneously adding a pair to the condensate in the superconducting metal. If the electron carries $ev_F$ of current ($v_F$ is the Fermi velocity), then $2ev_F$ of net current is flowing because the re-

 

turning hole carries $(-e)(-v_F)$. Of course, to conserve current, the electron pair also carries $2ev_F$. At low voltage, $eV<\Delta$, all of the electrons impinge upon the gap and Andreev reflect, so that twice as much current flows as in the normal state. In other words, the differential resistance is $R_0/2$. This result should not be too surprising. Half of the normal-state resistance comes from each metal, and in the limit of no applied voltage, the superconducting side is in equilibrium and thus resistanceless.

At voltages much greater than the gap only some fraction of the incident electrons, proportional to $\Delta/eV$, carry twice the normal current. The remaining electrons pass through essentially unimpeded by the gap, each contributing $ev_F$ to the current. Thus, we may write, for $eV \gg \Delta$,

$$I \propto ev_F(eV-\Delta)+2ev_F\Delta \approx \frac{V}{R_0}+\frac{\Delta}{eR_0} ,$$

where the first term comes from electrons above the gap, and the second from electrons below. The latter term is known as the "excess current," and is an exact result providing the gap rises from zero to its bulk value over a distance longer than the coherence length $\xi$. In this regime there is no Andreev reflection for $E>\Delta$. However, in a three-dimensional microconstriction the gap rises on a length scale comparable to $a$, which is often much shorter than $\xi$. As is often true in quantum mechanics, a "sharp-edged" barrier produces partial reflections for energies outside the classically forbidden region. These partial reflections also contribute to the excess current, yielding a value $\frac{1}{3}$ larger than above, i.e., $4\Delta/3eR_0$. The full $I$-$V$ curve in the pure metallic limit can be seen in Fig. 1, as the curve labeled $Z=0$.

If there is a tunnel barrier at the interface, electrons undergo some normal reflection there as well as Andreev reflection and transmission. As the strength of the barrier is increased, normal reflection begins to dominate, the excess current (proportional to the fraction Andreev reflected) gradually disappears, and the $I$-$V$ curve becomes more like a tunnel junction. This sequence is shown in Fig. 1, where $Z$ is a dimensionless parameter which measures the barrier strength. $Z$ has a simple interpretation; in the normal state, or at high voltages, the resistance is $R_n=R_0(1+Z^2)$.

Although $Z$ can be related directly to the "classic," square oxide-barrier transmission coefficient, we avoid making so specific a correspondence. Instead, $Z$ should be thought of as a phenomenological parameter to measure the elastic scattering in the neck, whether it originates from dislocations, oxide, or surface irregularities. In addition, $Z$ must remain a phenomenological parameter because our calculations have been performed for a one-dimensional system, yet they have been applied to a three-dimensional microconstriction.

A barrier is not the only source for normal reflection. In a more realistic system the Fermi velocities will be different in each bank. This "impedance" mismatch will result in some normal reflection, even with no barrier present. Figure 2 illustrates a typical result for a pure metallic $I$-$V$ curve, when the Fermi velocity ratio $r=\sqrt{2}$. One can easily show[5] that this effect is exactly reproduced by simply shifting $Z$ to a higher effective value,

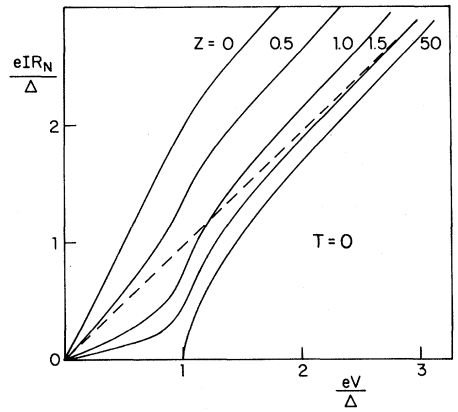


FIG. 1. Family of $I$-$V$ curves at $T=0$, where $Z$ is a measure of the interfacial scattering (see text). $Z=0$ curve corresponds to a pure metallic bridge, while $Z \gg 1$ corresponds to a tunnel junction. Dotted line is the normal-state $I$-$V$ curve. (This figure is reproduced from Ref. 1.)

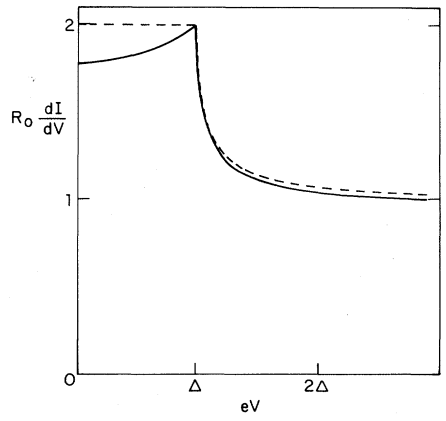


FIG. 2. $dI/dV$ vs $V$ at $T=0$ for the pure metallic ($Z=0$) case. Dotted curve assumes the Fermi velocities are equal in both metals; solid curve assumes the ratio $r$ of Fermi velocities is equal to $\sqrt{2}$. Latter curve is mathematically equivalent to one generated assuming equal Fermi velocities and $Z=0.175$.

$$Z_{\text{eff}}=[Z^2+(1-r)^2/4r]^{1/2},$$

while setting the Fermi velocities equal. Figure 3 plots $Z_{\text{eff}}$ vs $Z$, as a function of $r$, including a case appropriate for Cu-Nb. In view of this result, and of the doubtful validity of using a single parameter ($v_F$) to capture the complexities of the Fermi surface, in what follows the impedance mismatch is included simply as a shift in the measured $Z$.

## III. SAMPLE PREPARATION

The point contacts were formed by pressing the tip of a Nb wire against a Cu disk. Since a long mean free path is required to be in the Sharvin limit and to assure good electronic cooling, we used copper that was 99.999% pure. The Nb wire was of a lower purity, and had a resistivity ratio of $\rho_{300\,\text{K}}/\rho_{10\,\text{K}}=6$. From the $\rho l$ product of Nb ($4.7\times10^{-12}$ $\Omega$ cm$^2$) (Ref. 6) and our measured $\rho(10\ \text{K})=3\times10^{-6}$ $\Omega$ cm, we find a mean free path $\sim150$ Å. Cooling should be adequate for hole radii less than the mean free path. Similarly, our model (as described in Ref. 1) remains valid only in the ballistic limit, and thus we require $a<150$ Å in these metals.

The surface of the disk was mechanically polished until a specular finish was obtained. Then an etching solution of $HNO_3$:HF: acetic acid in a 5:4:1 ratio by volume was used to chemically polish the surface. This process produced a slightly rough, clean surface which was more desirable than the polished surface. We found experimentally that a polished surface produced less stable points, and with less variety in $Z$, than did the etched surface.

The 75-$\mu$m-diameter Nb wire was electrochemically sharpened to a point of radius less than 1 $\mu$m, using the same acid as above. A 5-V dc potential between the wire and the acid bath resulted in a smooth metal finish. Points exhibiting the greatest range of $I$-$V$ curve characteristics were slightly more concave than a simple cone shape, and had a length approximately 3 times longer than the wire diameter.

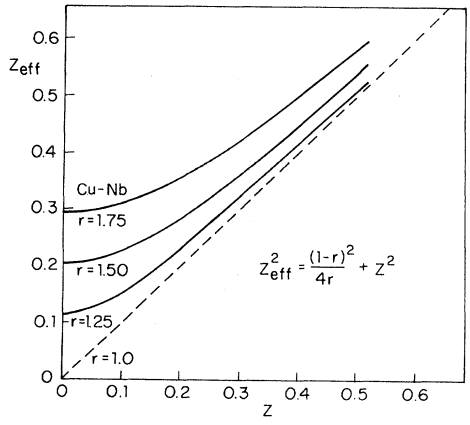


FIG. 3. $Z_{\text{eff}}$ vs $Z$, for a variety of Fermi velocity ratios $r$. Here, the $\delta$-function barrier is $V(x)=H\delta(x)$, $Z=mH/\hbar^2\sqrt{v_1v_2}$, $r=v_1/v_2$, and the $v$'s are the Fermi velocities in the two metals. Note that the result for $Z_{\text{eff}}$ is the same if the Fermi velocities are interchanged.

Both the wire and the flat are covered by a native oxide produced by exposure to laboratory air and the chemical etch. For the copper, this is the semiconducting oxide $Cu_2O$ which grows almost immediately to about 10 Å, and then stops.[7,8] In the niobium, the situation is more complicated[9–12] and perhaps still not completely understood, especially after wet chemical etching. It is thought to be as follows. The first few angstroms near the surface is metallic NbO, a superconductor with $T_c=1.4$ K. Above this layer is a semiconducting, microcrystalline amorphous layer of $Nb_2O_5$. Surface states, hybridization of the $d$ electrons, and adsorbed $H_2O$ all contribute to spatial variation of the energy barrier in the niobium oxide.[12] The effective barrier increases in height away from the bulk Nb, peaking just inside the topmost oxide layer. The first 20 Å (nearest the NbO) has a high density of states ($\sim10^{20}$/cm$^3$) and is basically metallic in behavior. A total of approximately 60 Å of oxide would be present after chemical etching. Both oxides are expected to be abraded by the action of touching the point to the flat.

The pointed wire and the Cu flat were mounted in the low-temperature rig, and immersed in liquid helium. Mechanical feedthroughs and a differential screw allowed stable positioning of the point. Since our major concern was to produce a variety of $I$-$V$ curves, accuracy of positioning was less important than stability against vibration.

In principle, the theory could be tested by measuring the $I$-$V$ curve as a function of temperature. Unfortunately, as the temperature is swept the point pressure will sporadically vary due to thermal expansion of the rig, thus changing $a$ in an unpredictable manner.[13] However, we are able to test the $Z$ dependence at low temperatures where the gap structure is the sharpest.

The lowest temperature achieved by pumping on liquid helium corresponds to $T/T_c=0.138$. At this temperature mechanical contact between the point and the disk was made, and the $I$-$V$ curve was displayed on an oscilloscope. Simultaneously, the curve was digitally sampled and stored on an LSI/11 computer for later analysis. The contacts were current biased using a battery-operated ramp. Isolation and noise filtering between the sample and the computer were provided by two PAR 113 preamplifiers.

## IV. EXPERIMENTAL RESULTS

Point contacts exhibit a seemingly endless variety of $I$-$V$ curve shapes. Thus, it appears to be difficult to codify this complexity into variations on a few simple themes, although many schemes have been proposed.[14] However, if much of this variation is due to changes in $Z_{\text{eff}}$, then some simplicity in organization may be possible. We have found that in a typical experimental run, the $I$-$V$ curves progress through three distinct regions with qualitatively different shapes. We argue below that these regions are distinguished by the length scale $s$, over which the electric field exists, compared to the oxide thickness $t$, hole size $a$, or the mean free path $l_{\text{Nb}}$. This length scale $s$ is inferred from our knowledge of the oxide characteristics and of the conduction processes in small constrictions. The regions may also be experimentally delineated via the normal-state resistance, into region I ($R > 100\ \Omega$), region II ($1\ \Omega < R < 100\ \Omega$), and region III ($R < 1\ \Omega$) (see Fig. 4).

### A. Region I: [$s < t, R > 100\ \Omega$]

After making the initial contact, the resistance is generally found to be above 100 Ω. The $I$-$V$ curves are observed to be linear, showing no gap structure whatsoever. This unexpected behavior suggests that we are dealing with an $N$-$N$ microconstriction rather than an $N$-$S$ one. We are thus led to try to understand the conditions under which the superconductivity of the niobium can be kept from playing a role in the $I$-$V$ curve.

The model we suggest is the following: Imagine that some dirty normal metal coats the surface of the Nb so that the Cu-Nb contact can be thought of as a Cu—oxide barrier—normal metal—Nb sandwich. In this case, the electric field will be centered in the resistive interfacial oxide and contained entirely between the normal-metal conductors. The dirty normal layer will assure enough scattering to decouple the electrons in the region over which the voltage drop appears, from the proximity effect of the superconducting Nb electrode. Consequently, the $I$-$V$ curve will be structureless and linear.

We suggest that this model is appropriate to our region-I contacts because, as explained in the section above, the first 20 Å of niobium oxide is a semiconductor with a large density of interface states. This material should to a good approximation act as a dirty normal layer [see Fig. 4(a)]. Providing the electric field remains in the normal region (i.e., $s < t$), we expect a linear $I$-$V$ curve. This latter requirement can be met if the contact area has a radius less than 10 Å (as is often true in region II below), or if the oxide barrier is high. In either case, the electric field will be constrained to be within the oxide. Most likely, both possibilities combine to produce the observed curves. However, the oxide layers are easily removed by the mechanical movement of the point. Eventually, with repeated making and breaking of contact, these high-resistance $I$-

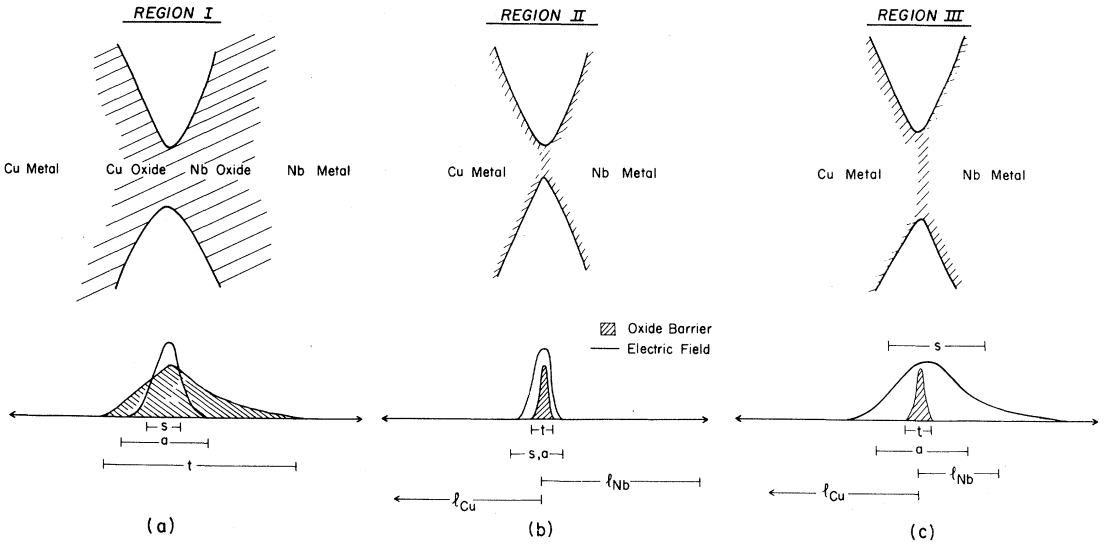


FIG. 4. Inferred contact geometries and electric field distribution. (a) region I. The electric field is contained entirely within the thick, normal metallic oxide layer, leading to a purely linear $I$-$V$ curve. (b) region II. The electric field penetrates into each bank a distance $\sim a$, the hole radius. The mean free path in either bank is greater than $a$, i.e., within the Sharvin limit. Most $I$-$V$ curves can be fit with a $Z_{\text{eff}}$ parameter. (c) region III. $l_{\text{Nb}} < a$. The electric field penetrates a distance greater than $l$ into the Nb, and the current flow is diffusive. $I$-$V$ curve shows clear evidence of heating.

$V$ curves are less frequently observed, and region I gives way to the lower-resistance region II.

### B. Region II: $[t < a < l_{Nb}, 1\ \Omega < R < 100\ \Omega]$

In this resistance regime, the thick oxide has been abraded and the hole radius, as inferred from the measured resistance and the $\rho l$ products of Cu (Ref. 15) and Nb,[6] varies from 10 to 120 Å. This is the Sharvin limit ($l > a$), and the electric field penetrates into each bank a distance comparable to $a$ [Fig. 4(b)]. Distinct gap structure is seen, and nearly all the $I$-$V$ curves can be accounted for by our theory. The sole exception is when the $I$-$V$ curve shows structure at $\sim\Delta_{Nb}/6$, which we believe is due to a gap in NbO of roughly that magnitude.[16] Such structure is occasionally seen, as illustrated by Fig. 5.

Figures 6 and 7 show a variety of more typical $I$-$V$ curves, along with a fit to theory. Over 30 curves have been examined, of which these are a *representative* sample. The voltage scale is fixed by the literature value for the gap (1.47 meV).[17] [This is the gap value appropriate for oxygen-rich (i.e., dirty) niobium, as expected, given the low-resistivity ratio of our bulk Nb wire.] Thus, the plots shown are two parameter fits: We measure the normal-state resistance $R_n$ to set the current scale, and then adjust the barrier height $Z_{eff}$. Note that the lowest $Z_{eff}$ value observed is 0.3, in good agreement with the impedance mismatch effect discussed above (see Fig. 3). $Z_{eff}$ values are rarely observed above $\sim 1$, an indication that in region II the oxide is almost entirely flaked off by the point. A high-quality fit is routinely seen and one can generally distinguish by eye

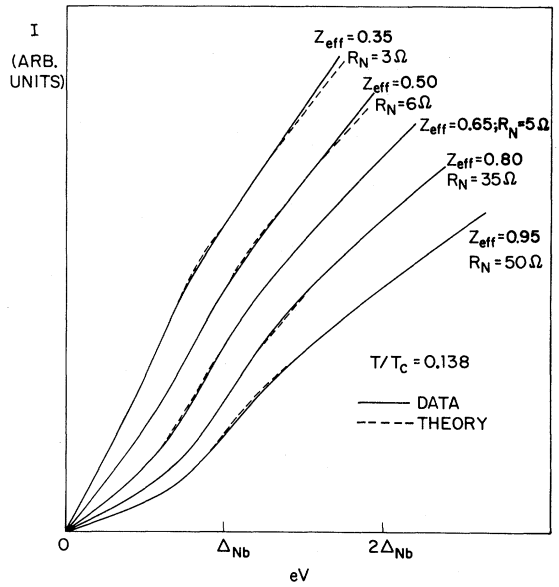


FIG. 6. Region-II $I$-$V$ curves at $T/T_c = 0.138$. Solid lines are the experimental results, dotted lines are the fit to theory. Scaling of the current axis is roughly in units of $\Delta/eR_0$, but selected in each case so as to prevent crowding of the curves. Where the experimental and theoretical curves overlap, only the experimental result is shown.

a best-fit $Z_{eff}$ value within $\pm 0.025$.

We have also observed the separate (i.e., multiplicative) contributions to the normal-state resistance of the geometric resistance ($R_0$) and of the barrier ($1+Z_{eff}^2$). By gently adjusting the point pressure while keeping the point fixed in one spot along the

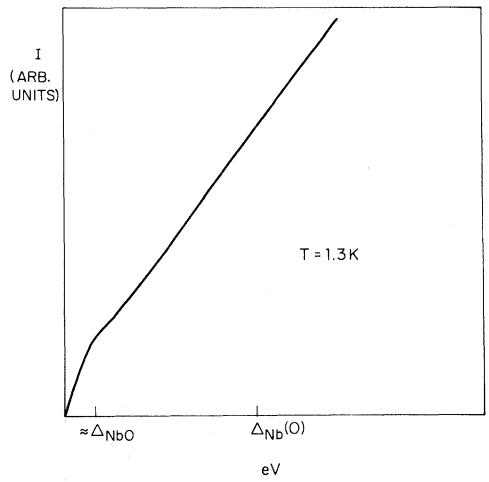


FIG. 5. Anomalous $I$-$V$ curve, showing the small-gap structure attributed to a superconducting NbO oxide layer.

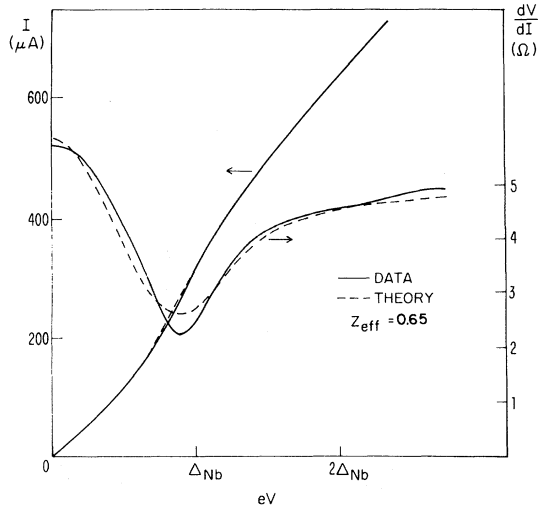


FIG. 7. Comparison of the theoretical and experimental curves of $dV/dI$ and $I$ vs $V$ for the case $Z_{eff} = 0.65$. This quality of fit is routinely observed.

surface, it is possible to obtain a family of $I$-$V$ curves varying in resistance by nearly 2 orders of magnitude, yet having fitted $Z_{\text{eff}}$ values ($Z_{\text{eff}} < 1$) clustered within $\pm 0.05$ of each other. Since $R_n = R_0(1+Z_{\text{eff}}^2)$, there is no way such a large variation in resistance can be attributed to such a small change in $Z_{\text{eff}}$. Rather, we believe that in this case the area of the constriction is varied while $Z_{\text{eff}}$ is essentially unchanged. This rather unexpected result can be explained as follows: Comparison with the classic rectangular oxide-barrier model shows that $Z \approx \sinh t$, where $t$ is the thickness of the oxide barrier in angstroms. Thus, for $Z < 1$, $Z$ depends only linearly (i.e., not exponentially) on the thickness $t$. These low $Z$ values correspond to only a monolayer or so of oxide, and further pressure may simply flatten the point, giving a larger contact area over the same minimal thickness of a tenacious oxide.

### C. Region III: $[a > l_{\text{Nb}}, a > t, R < 1\ \Omega]$

After manipulating the point a large number of times, the end begins to curl upon itself, as shown in Fig. 8, progressively widening the area of contact. This widening is not simply related to the nominal contact area, because the actual contact occurs only at small "high spots." Still, we suppose that the hole size of the actual contact is comparable to the niobium mean free path, so that current flow is now diffusive in the Nb, although the copper may still be in the Sharvin limit [see Fig. 4(c)]. Although the curve can roughly be assigned a $Z_{\text{eff}}$ value, it is clearly distorted by heating. Specifically, the fit requires a gap that is reduced from the bulk value, and a rising background can be seen in the derivative (Fig. 9). At this stage this point is discarded, and a new one is mounted.

## V. CONCLUSIONS

We have studied Cu-Nb point contacts as a model system for the $N$-$S$ microconstriction. By varying the pressure and position of the point we can generate $I$-$V$ curves ranging from metallic to tunnel junction behavior. In an earlier paper,[1] specific pre-

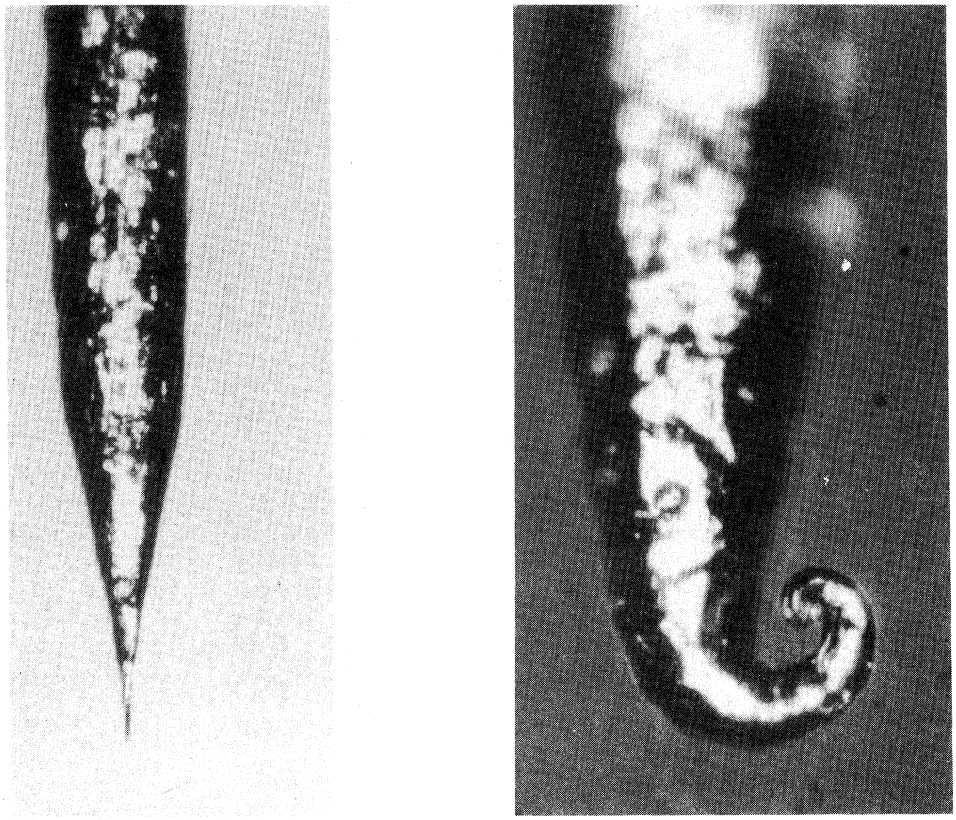


FIG. 8. (a) 75-$\mu$m-diameter wire after electrochemical etching. (b) Same wire after repeatedly touching the Cu flat until region-III $I$-$V$ curves were obtained.

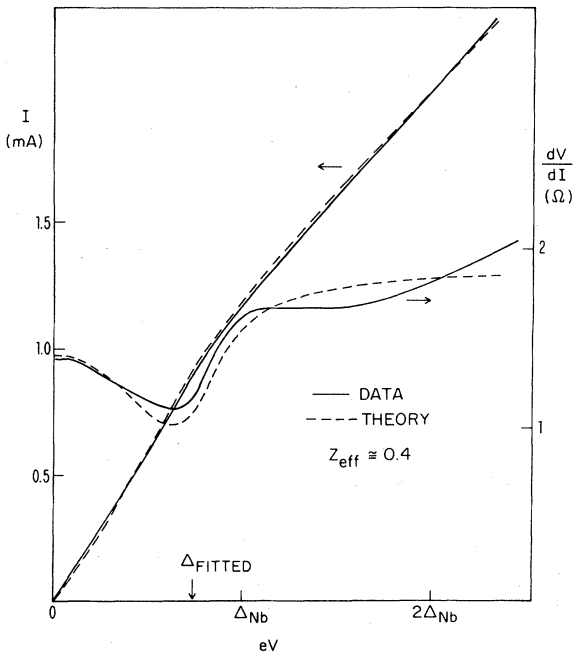


FIG. 9. Region-III *I-V* curve showing clear heating distortion. Specifically, the gap is lowered from the bulk value, and there is a rising background in the derivative. Even with Δ as an adjustable fitting parameter, the best fit of $Z_{eff}=0.4$ is rather poor.

dictions for the details of the crossover were presented. Providing that the electron transport remains in the Sharvin limit (region II), we find excellent agreement between theory and experiment. Two other regimes, one dominated by heating (region III) and one by oxide conduction (region I), were also found.

A number of simplifying assumptions were necessary to translate the basic physics of our model into specific *I-V* curve predictions. Still, the experimental results indicate that the gap structure and excess current at an *N-S* microconstriction are primarily determined by Andreev reflection, and that this process is adequately described by our simple model. This approach should also give insight into the detailed behavior of high-current-density tunnel junctions, microbridges, and dielectric breakdown sandwiches.

## ACKNOWLEDGMENTS

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[14]For example, a parallel combination of tunnel junction and a microbridge has been proposed to explain the *I-V* curves of *S-S* point contacts (Yu. Ya. Divin and F. Ya. Nad', Fiz. Nizk. Temp. 4, 1105 (1978) [Sov. J. Low Temp. Phys. 4, 520 (1979)]). Competition between the oxide thickness $t$ and the hole radius $a$ has been used in explaining noise in semiconducting point contacts [L. K. J. Vandamme, J. Appl. Phys. 45, 4563 (1974)]. Generally, however, experiments have focused more on the device applications of point contacts than on the basic physics behind experimentally accessible *I-V* curves.

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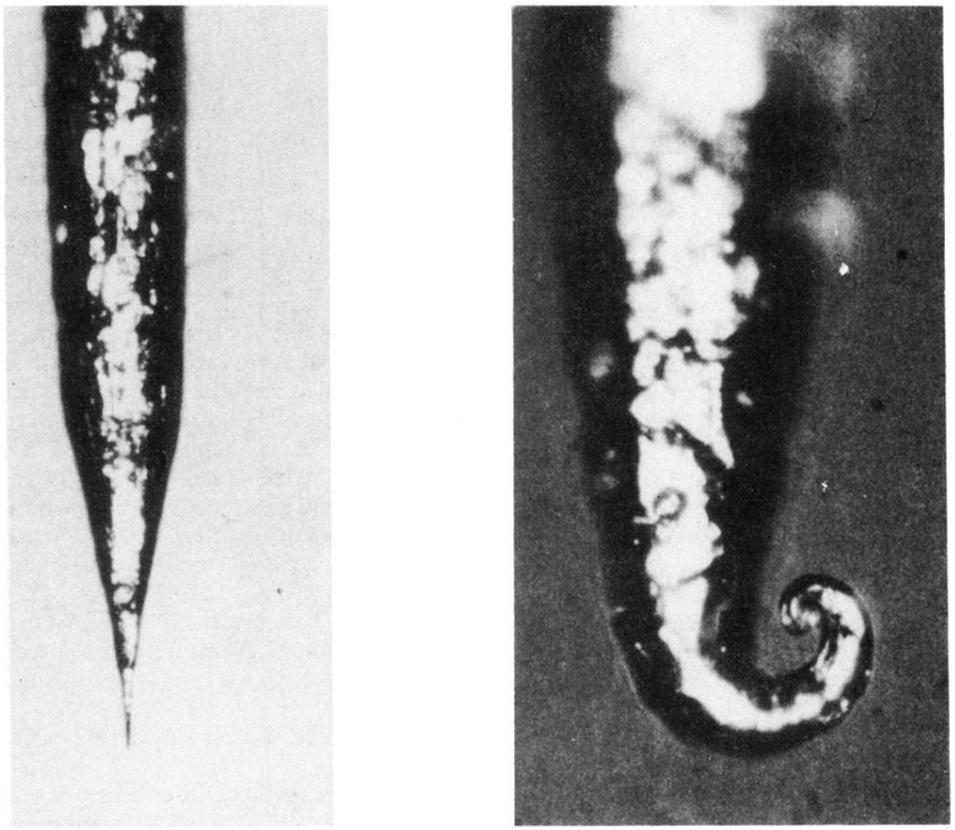


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