# Single-atom point contacts in the superconducting state

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Recently it has become possible to fabricate long-term stable point contacts with a constriction as small as a single atom. Here we present experimental investigations on one-atom point contacts in the superconducting state. An applied pressure or tensile force on this atom results in large effects in the superconducting properties. Furthermore, we show that one-atom point contacts can be broken and reestablished in a perfectly reproducible manner.

#### **INTRODUCTION**

Inherently stable one-atom point contacts can be realized experimentally fairly simple. Various properties of these smallest possible contacts between two electrodes were studied experimentally and it was found that the conductance carried by one atom is of the order of  $2e^2/h$ . However, a number of fundamental issues related to these microscopic contacts are still unknown in part because of a lack of theories describing these systems. A related difficulty is the nonuniversality, the "sample to sample" fluctuations. For example, two one-atom point contacts of the same material may show a difference in conductance reflecting a difference in a nonadiabaticity of the potential distribution in the contact region. This in term may result from a defect or a difference in electrode geometry coupled to the bridging atom. On the other hand it has been shown for a material like copper that various one atom contacts show a conduction close to  $2e^2/h$  (see Ref. 1), indicating that up to some extent there exists a universality with a statistical mean which can be calculated. Indeed the theoretical simulations from Todorov and Sutton<sup>2</sup> have a nice resemblance to the experiment in this particular case.

In this work we present experiments performed on single-atom point contacts in the superconducting state. We focus on effects at the single-atom level which are reproducible, we do not attempt to obtain a statistical mean over many different single-atom point contacts.

## **EXPERIMENTS**

We used the mechanically controllable break (MCB) technique to obtain atomic size contacts.<sup>3</sup> A schematic drawing of the MCB sample mounting is shown in the upper inset of Fig. 1. The basic principle of this technique is that a length change of the piezo results in a reduced change of the separation between the two electrode halves. The reduction factor in our samples, imposed by the sample geometry, is estimated to be of the order of  $10^2$  to  $10^3$ . Due to this large factor it is possible to change contact sizes over subatomic dimensions, enabling us to study, e.g., contact formation or to fabricate inherently stable contacts with a constriction as small as a single atom. This technique utilizes the roughness on an atomic scale of two freshly broken electrodes. The experiments are performed at 1.3 K, magnetic shielding ( $\mu$ 

metal) yields a field at the sample location less than 1  $\mu$ T. All measurements leads entering the cryostat are filtered using feedthrough  $\pi$  filters. In addition, before entering the copper and lead shielded sample area the leads are filtered by low-temperature copper powder filters. The experimental setup is the same as described in Ref. 3. All experiments are performed by current biasing the sample in a standard four-probe configuration.

In Fig. 1 a current-voltage (I-V) curve is shown of a contact which consists of one atom. The I-V curve shows a clear linear part for voltages larger than the superconducting gap voltage  $2\Delta/e$ . The slope of this linear part is used to determine the normal resistance,  $R_N$ , of the contact. Sharp kinks in the voltage range  $\pm 2\Delta/e$  at voltages  $\pm 2\Delta/ne$  with  $n = 1, 2, 3, \ldots$  are present. This subgap structure is observable over a wide range of  $R_N$  values. Subgap structure is known to be present in I-V curves of point contacts, however also tunnel junctions show this kind of structure.<sup>4</sup> Near zero voltage a clear



FIG. 1. The *I-V* curve of a single-atom contact (Sn electrodes) shows the threshold current region, subgap structure, and a linear branch. The lower inset is an enlargement of the central section, the arrow indicates the threshold current  $I_i$ . The upper inset shows a schematic drawing of the sample mounting, with A: bending beam, B: counter support, C: notched metal filament, D: epoxy adhesive and E: piezo element.

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finite critical current is present. In Ref. 5 it was shown that in the tunnel regime and in the high Ohmic contact regime, as shown here, this critical current obtained a small slope. This is more clearly visible in the lower inset, which is an enlargement of the central section. The critical current exhibits a small slope, which is not an artifact of the four probe measurement. Because the I-Vcurve does not show a zero-voltage critical current the maximum current, indicated by the arrow in Fig. 1, was called a threshold current  $I_t$ . It was speculated that the threshold current originates from Cooper pairs which gain enough energy from the environment to overcome a Coulomb barrier. In correspondence with Ref. 5 we will also use the  $I_t$  notation here for the "sloped supercurrent". When increasing the current in the I-V curve of Fig. 1 there is a sudden jump to a large voltage (300  $\mu$ V) whenever the current reaches  $I_t$ . This is characteristic for all our I-V curves of superconductors. Here this jump is used to define  $I_t$  experimentally by using a setpoint voltage of typically 100  $\mu$ V. The equipment ramps the current; when the setpoint voltage is reached the current switches back to zero. The current value at the setpoint is held at an output terminal and is updated with a frequency of 20 Hz. This procedure is used in the following experiments to continuously monitor  $I_t$  when changing the contact configuration over subatomic distances. The normal resistance is determined with a lockin technique at a current level with corresponding voltages much larger than  $2\Delta/e$ .

In the following experiments the transitions from contact to tunneling and tunneling to contact are studied for the two superconductors Sn and Ta. Only the data in the contact regime will be displayed. The first stage of the ex experiment, a one-atom contact, is manually adjusted with the use of a voltage on the piezo. Once this is achieved a small slowly varying piezo voltage ramp  $(V_p)$ is superimposed on the voltage. This triangularly shaped ramp is shown in the top panel of Fig. 2. The next panel shows the continuous  $I_t$  measurement of a Sn junction during the  $V_p$  variation. At t=0 the junction is in the tunnel regime. By reducing the piezo voltage the two electrodes will come closer together. Because the electrodes are broken prior to operation they are rough on an atomic scale and a single-atom contact will be formed where two atoms on opposite electrodes are nearest. At a certain point a jump to contact is observed, which results in a relatively large  $I_t$  value of 25 nA. Reducing  $V_p$  even further results in a changing pressure at the contact location and an increase of  $I_t$ . When increasing  $V_p$  (pulling the electrodes apart),  $I_t$  decreases even beyond the 25-nA value. Then a step in  $I_t$  occurs to approximately its double value. We attribute this jump to a sudden rearrangement of the atom, which bridges the electrodes. When the electrodes are pulled further apart  $I_t$  decreases again, followed by a jump to the tunnel regime where tunneling takes over. In this regime the equipment is not capable of measuring  $I_t$  continuously. However, from I-V curves taken in this regime we know that  $I_t$  is typically smaller than 1 nA. Repeating the  $V_p$  sweep leads to a perfectly reproducible measurement showing that it is not only possible to make a single-atom contact, but also that applied changes of pressure on this single atom result in reproducible effects.

This reproducibility allows us to perform a continuous  $R_N$  measurement after the  $I_t$  sequence is recorded. Thus, although the  $I_t$  and  $R_N$  measurements are not performed at the same time, they can be attributed to exactly the same contact configuration. The  $R_N$  measurement is shown in the third panel of Fig. 2. The maximum in the piezo voltage served as a zero time reference, so at a time t the  $I_t$  and  $R_N$  panel show the corresponding values for a specific contact configuration. Coming from the tunnel



FIG. 2. The nature of the experiment is to break and reestablish a one-atom contact; this is achieved with Sn by applying a slowly varying triangular voltage on the piezo element (upper panel). The rest of the panels display data in the contact regime only. Starting in the tunnel regime (t=0) the reduction of  $V_p$ (electrodes move towards each other) results in a one-atom contact. A subsequent increase in  $V_p$  (electrodes move apart) results in a rearrangement of the atom followed by a jump back into the tunnel regime. Both the  $I_t$  and  $R_N$  measurement represent the same contact at a specific time t. Furthermore, the jump to contact, the rearrangement of the atom and the jump into the tunnel regime line up perfectly in the  $I_t$  and  $R_N$ panel. The arrows indicate the subsequent increase or decrease of  $I_t$  and  $R_N$  after a jump. The reproducibility shows that during contact formation and breaking the bridging atom favors one specific site. The  $I_t R_N$  panel is obtained by multiplying the  $I_t$  with the  $R_N$  panel.

regime  $(R_N \text{ typically larger than 40 k}\Omega)$  a jump to a 6 $k\Omega$  contact value is measured simultaneously with the discontinuous  $I_t$  increase. Initially  $R_N$  decreases with increasing pressure and  $R_N$  increases when the tips are pulled apart. The jump to another point-contact configuration lines up perfectly with the jump observed in  $I_t$ . However, the relative size of the jump is about 25% for  $R_N$  whereas  $I_t$  shows an increase of about 100%. After this rearrangement  $R_N$  decreases and slightly increases. A clear turnup is visible just before a jump to the tunnel regime occurs. During this trajectory  $I_t$  shows a monotonic decrease. The  $I_t R_N$  result, obtained by multiplying the  $I_t$  panel with the  $R_N$  panel, is shown in the bottom panel of Fig. 2. This panel mainly reflects the dependence of  $I_t$  on  $V_p$  due to the much larger variations in  $I_t$  than in  $R_N$ .

A similar measurement is performed on a Ta break junction, which is shown in Fig. 3. The  $I_{i}$  panel shows a variation of approximately a factor of 2 in  $I_t$  with changing  $V_p$ . As in the Sn measurement there is a jump to tunneling for increasing  $V_p$  followed by a jump to contact when  $V_p$  is reduced. In this measurement, however, we do not observe a jump in the contact regime, which means that the constriction atom does not rearrange with respect to both electrodes. The measurement is again

7.5 0.2  $_{t}R_{N}(mV)$ 0.1 0.0 0 200 400 600 t(s) FIG. 3. A similar measurement as presented in Fig. 2 performed on superconducting Ta. Note the factor of 2 change in

 $I_t$  during the continuous increase of  $R_N$ .

perfectly reproducible. The  $R_N$  panel shows a  $R_N$  variation of only 7% in the contact regime. For decreasing  $V_p$ (increasing contact pressure) a reduction of  $R_N$  coincides with a reduction in  $I_t$ . For increasing  $V_p$  (pulling the electrodes apart) a monotonic increase of  $R_N$  coincides with an increase followed by a decrease of  $I_r$  until the jump to tunneling occurs. Similar as in Fig. 2 the  $I_t R_N$ panel is reflecting mainly the  $I_t$  variations (of almost a factor of 2).

### DISCUSSION

Both measurements show a hysteresis in contact formation and the breaking of the contact. The jump to contact takes place at a smaller  $V_p$  than the jump to the tunnel regime. This hysteresis reflects the elasticity in the electrode ends and the bonding forces of the front most atoms once a contact is formed. When a one-atom contact is formed a higher  $V_p$  is needed to break the contact due to some elasticity in the electrode ends. The value of the resistance when a contact is formed or is broken is difficult to predict. The Sharvin resistance,<sup>6</sup> which relates resistance to constriction diameter, is not valid for contacts of the order of the Fermi wavelength. The Landauer formalism provides a conductance of  $2e^2/h$  per conductance channel. When we attribute one conductance channel to one atom, we expect a resistance of  $h/2e^2 \simeq 13 \text{ k}\Omega$  for a single-atom contact. From 20 measurements similar as shown in Figs. 2 and 3, performed with superconducting electrode material consisting of Sn, Ta, Nb, In, and Pb, we conclude that the jump to tunneling for these materials occurred at a conductance value between  $2e^2/h$  and  $4e^2/h$ .

It is difficult to predict how thermal fluctuations will effect the  $I_t$  value. At 1.3 K, using the Josephson coupling energy,  $k_B T = \hbar I_T / 2e$  results in a thermal current,  $I_T$ , of 55 nA which is comparable to the measured threshold current. In our case zero-point fluctuations in the parabolic part of the washboard potential may play an even more important role than thermal fluctuations. Assuming a capacitance between  $10^{-14}$  and  $10^{-15}$  F for the single-atom contact, the zero-point energy  $\frac{1}{2}\hbar\omega_p$  is larger then  $k_B T$  and is comparable to the Josephson coupling energy, which equals the amplitude of the washboard potential. However, at this stage we are not able to describe the interplay between the thermal fluctuations, the zero-point fluctuations and the Coulomb barrier associated with the small capacitance. It is shown experimentally that the  $I_t$  variation can be as large as a factor of 2, whereas the normal resistance undergoes a relatively small variation. This leads to a large variation of the  $I_t R_N$  product. In contrast to this finding, in tunnel junctions the  $I_t R_N$  product was found to be approximately constant.<sup>5</sup> The explanation for this result may find its origin in the scattering on a single defect in the contact region<sup>7</sup> and the change in scattering probabilities when a pressure or a tensile force is applied on the atom which spans the electrodes.

In conclusion we have examined the superconducting characteristics of one-atom point contacts. It is shown



atom.

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- <sup>1</sup>J. M. Krans, C. J. Muller, I. K. Yanson, Th. C. M Govaert, R. Hesper, and J. M. van Ruitenbeek, Phys. Rev. B 48, 14721 (1993).
- <sup>2</sup>T. N. Todorov and A. P. Sutton, Phys. Rev. Lett. **70**, 2138 (1993).
- <sup>3</sup>C. J. Muller, M. C. Koops, B. J. Vleeming, R. de Bruyn Ouboter, and A. N. Omelyanchouk, Physica C 220, 258 (1994).
- <sup>4</sup>Subgap structure in planar-oxide tunnel junctions has often been attributed to microshorts penetrating the barrier region.

From the MCB experiments it is clear that vacuum barrier tunnel junctions also show this structure see, e.g., N. van der Post, E. T. Peters, I. K. Yanson, and J. M. van Ruitenbeek, Phys. Rev. Lett. 73, 2611 (1994).

- <sup>5</sup>C. J. Muller and R. de Bruyn Ouboter, Proceedings of the 20th International Conference on Low Temperature Physics [Physica B 194-196, 1043 (1994).
- <sup>6</sup>Yu. V. Sharvin, Zh. Eksp. Teor. Fiz. **48**, 984 (1965) [Sov. Phys. JETP **21**, 655 (1965)].
- <sup>7</sup>A. N. Omelyanchouk, R. de Bruyn Ouboter, and C. J. Muller, Low Temp. Phys. 20, 398 (1994).