Subgap Structure as Function of the Barrier in Atom-Size Superconducting Tunnel Junctions

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Using a mechanically controllable breakjunction we study superconducting tunnel junctions of atom size. The vacuum barrier is adjusted by controlling the distance between the electrodes to give barrier transparencies ranging over 2 orders of magnitude. We observe a systematic variation of the subgap current steps at $eV = 2\Delta/n$, for $n = 2$ and $n = 3$ in excellent agreement with theories of multiparticle tunneling and multiple Andreev reflection.

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In superconductor-insulator-superconductor (SIS) tunnel junctions the superconducting energy gap Δ is observed as a well-defined step in the current at bias $eV = 2\Delta$. This is the threshold energy for the transfer of single quasiparticles, and at zero temperature current vanishes below this bias. In 1963, Taylor and Burstein [1] were the first who observed current steps at $eV = 2\Delta/n\,(n = 1,2,\dots)$ in these junctions. Three mechanisms have been suggested to describe this phenomenon: multiparticle tunneling (MPT) [2,3], Josephson self-coupling (JSC) [4], and multiple Andreev reflection (MAR) [5]. The experimental observation of anomalies in the current at biases $eV = 2\Delta/n$ are reproduced in each of the theories. It is presently believed that JSC is not appropriate to describe subgap structure [5], and MPT and MAR are now regarded as describing the same mechanism in the two limits of low and high transparency, respectively [6,7]. Uncertainty about crucial junction parameters in the experiments on planar superconductor-insulator-superconductor tunnel junctions such as thickness, surface area, transparency, composition, and dimensionality of the barrier has prohibited quantitative tests of the theories. Recently, interest in the phenomena revived, stimulated by the possible application of these junctions as x-ray detectors [8,9].

In this Letter we present a new approach, where a mechanically controllable breakjunction (MCB) is used to continuously adjust the width of the tunneling barrier. This system has none of the drawbacks of planar tunnel junctions: tunneling takes place through a true vacuum barrier, and the current passes through the frontmost atoms of the junction. The experiments show that the observed subgap structure is in excellent agreement with the predictions of MPT and MAR.

The MCB technique (cf. Fig. 1) [10] uses a (superconducting) metal wire which is glued on a substrate and can be broken by bending the substrate. Breaking the wire at low temperature and high vacuum guarantees two atomically clean surfaces. The surfaces can be brought together again, and the distance can be controlled by a piezoelectric element. Reducing the piezovoltage, the surfaces will be pressed together and a point contact with resistance less than an ohm can be formed. Increasing the piezovoltage results in a smaller point contact, which is eventually reduced to only a few atoms. In this regime the resistance increases stepwise, since the contact breaks down atom by atom. It is possible to form a point contact consisting of only a single atom [11]. The difference between a one atom MCB junction and a one atom STM junction is the exceptional stability of the former (better than 10^{-13} m), allowing detailed current-voltage measurements. The resistance R_c of a one atom contact depends on the material and has a value close to the quantum resistance $h/2e^2 \simeq 12.9\ \mathrm{k}\Omega$ [12]. For the materials used in this study (Pb and Nb), R_c lies between 6 and 11 kΩ. The relatively large spread in this value comes from variations in the geometry of the atoms around the contact. When the piezovoltage is increased still further, a jump to tunneling is observed and a vacuum barrier is formed between the two foremost atoms. In this case we have a single atom tunnel junction with a transparency T decreasing with the width d of the vacuum barrier [13].

We measured dc current voltage (I-V) characteristics of Nb and Pb tunnel junctions at 1.5 K using both current and voltage bias. A typical I-V curve of a Nb junction with $R_n = 145\ \mathrm{k}\Omega$ is depicted in Fig. 2. The superconducting gap is clearly visible at a bias of $2\Delta = 2.85$ meV. This value agrees with the literature values of 2Δ which are between 2.85 and 3.05 meV [14]. Visible in the inset are the subgap current steps dJ_n at biases $eV = 2\Delta/n$ for $n = 2$ and 3. The position of the steps,

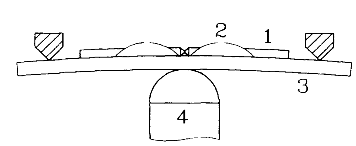

FIG. 1. Schematic picture of the breakjunction technique. A superconducting wire (1) is glued with epoxy adhesive (2) on a bending beam (3) and can be broken at low temperature and high vacuum, and a piezoelement (4) allows fine control of the distance between the two freshly exposed surfaces.

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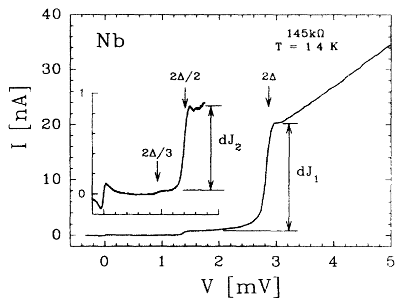

FIG. 2. Current-voltage characteristics of a Nb MCB junction with $R_n = 145$ kΩ, at 1.4 K. Subgap currents at $eV = 2\Delta/n$ are clearly visible on the expanded vertical scale of the inset.

defined as the position of the maximum in dI/dV, is constant for the various junction resistances, and dJ_n is found at $2\Delta/n$ within the experimental accuracy of 5 μV.

A true supercurrent is not observed in these junctions. As shown by Muller *et al.* [10,15], the supercurrent is gradually reduced below the value of Ambegoakar and Baratoff [16] as the resistance of a point contact is increased above 100 Ω. It extrapolates to zero at the transition to tunneling at about 10 kΩ. In the tunneling regime, a small skew Josephson-like current survives.

Many curves similar to those in Fig. 2 were measured for both Pb and Nb, for various settings of the vacuum barrier width d. The magnitude of the current steps at $2\Delta/n$ decreases rapidly with respect to the step at 2Δ, when the separation between the electrodes is increased. The systematic variation of the subgap structure is brought out by the plot in Fig. 3 where we present the ratio of subgap currents dJ_n/dJ_{n-1}, for $n = 2$ and $n = 3$, as a function of $1/R_n$. We normalized $1/R_n$ by the quantum resistance $h/2e^2$. Here, we anticipate that the current steps dJ_n scale with the transparency T of the tunnelbarrier as $dJ_n \sim T^n$. The ratio of the steps at $2\Delta/n$ and $2\Delta/(n-1)$ would therefore scale as $dJ_n/dJ_{n-1} \sim T$. The normal state resistance is expected to depend on the same transparency

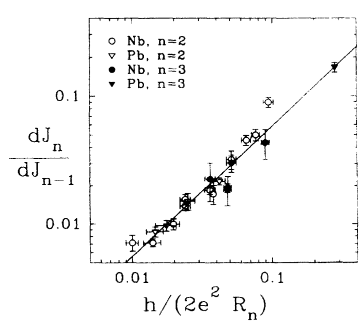

FIG. 3. Ratio of the subgap currents versus $h/2e^2R_n$, with R_n the normal resistance measured far above the gap and $h/2e^2$ the quantum resistance. The full line is a fit to the data points and corresponds to the prediction of MPT and MAR theories.

as $R_n \sim T^{-1}$. An example of the way in which dJ_n is extracted from the I-V curve is given in Fig. 2. R_n is determined from the slope of the line connecting points on the I-V curve at $eV = \pm 4\Delta$. Some nonlinearities in the I-V curve above the gap are taken into account and lead to small error bars. Figure 3 is given on a double logarithmic scale since we cover almost 2 orders of magnitude in R_n, i.e., R_n ranges from 45 kΩ to 1.3 MΩ. A least squares fit of the form $dJ_n/dJ_{n-1} = (R_0/R_n)^p$ gives $p = 1.00 \pm 0.05$, which is in excellent agreement with the expected dependence (see below). The constant R_0 is found to be $(0.61 \pm 0.06)\, h/2e^2$ or 7.9 ± 0.8 kΩ which is comparable to the value of a one atom contact. It is observed that for both Nb and Pb *both* dJ_2/dJ_1 and dJ_3/dJ_2 follow the same law. This is the first time that such systematic dependence is observed. In particular, we have $dJ_3/dJ_2 = dJ_2/dJ_1$ within the experimental accuracy. In previous experiments on planar junctions these ratios were found to disagree by many orders of magnitude as a result of the inhomogeneity of the barrier.

Among the explanations for the subharmonic structure, JSC is believed to be less probable. Based on our observations it can now be ruled out. JSC results from the reabsorption by a quasiparticle of n photons from the RF field generated by the Josephson current. However, in our junctions the supercurrent is strongly suppressed. As shown in Ref. [15] the product of the critical current and the normal resistance I_cR_n as a function of R_n decreases for R_n larger than 100 Ω in atom-size junctions. The product I_cR_n is effectively zero above 10 kΩ. Without the supercurrent there can be no Josephson radiation, and hence no JSC. The explanation for the suppression of I_c for very small junctions is believed to be either a pair breaking mechanism in ultrasmall junctions, or a size effect for junctions smaller than $a_L = v_F/\omega_D$ [15].

The two remaining mechanisms MAR and MPT describe essentially the same physics, as has been recognized by several authors [6,7]. Multiparticle tunneling [2,3] is the process of simultaneous tunneling of n quasiparticles across the barrier, each gaining an energy eV and combining to form Cooper pairs and (at most) one quasiparticle. The process is described by a tunneling Hamiltonian in lowest perturbation, and is thus applicable to cases where $T \ll 1$. The magnitude of the threshold currents dJ_n at $eV = 2\Delta/n$ is proportional to T^n so that $dJ_n/dJ_{n-1} \sim T$ [17]. For higher transparencies the perturbative approach breaks down and the subgap currents are more naturally described using MAR.

Andreev reflection is the process of an electron incident on the barrier converting into a backscattered hole upon creation of a Cooper pair. In MAR this process takes place n times, so that a quasiparticle can gain n times the bias eV making it possible to reach the threshold energy 2Δ. MAR has been used successfully to explain excess currents and subharmonic structure in microshorts [5,8] and planar superconductor-normal metal-superconductor

(SNS) junctions [18]. Just as in the case of MPT, n quasiparticles cross the barrier. The method for MAR used by Arnold [6] does not involve a perturbative tunneling Hamiltonian and is valid for arbitrary T, in contrast to MPT. According to Arnold, for SIS junctions the subgap structure disappears with decreasing T as $dJ_n/dJ_{n-1} \sim T$ for $T \ll 1$, just as MPT.

This prediction is in qualitative agreement with our measurements since we find $dJ_n/dJ_{n-1} = R_0/R_n$, and from basic tunneling theory [19] we expect $1/R_n \sim T$. The interpretation of the prefactor R_0 depends on the number of quantum channels that contribute to the tunnel current. For a tunnel junction, we view the potential barrier between the two front atoms to be squeezed forming a saddle point in the center of the junction so that the tunneling is dominated by a single channel. In this case the Landauer formula allows us to make the identification $T = R_Q/R_n$, with $R_Q = h/2e^2 = 12.9$ kΩ. With the experimental observation $dJ_n/dJ_{n-1} = R_0/R_n$ we obtain $dJ_n/dJ_{n-1} = (R_0/R_Q)T = (0.61 \pm 0.06)T$. The prefactor for MPT has not been calculated for quantum tip junctions. For MAR no detailed calculations of the current step sizes are available, only the order of the steps is predicted. Recently, Shumeiko and co-workers [20], using a model closely related to MAR, have calculated dJ_n for $T \ll 1$ and predict $dJ_n/dJ_{n-1} \simeq 0.5T$ for $n = 2$ and $n = 3$, in fair agreement with the above estimate. Using this approach we find that the transparency in our experiment ranges from 0.29 at $R_n = 45$ kΩ to 0.01 at 1.3 MΩ.

In summary, we have observed the theoretically predicted ratio and transparency dependence of the subgap current steps in atom-size superconducting tunnel junctions, for R_n between 45 kΩ to 1.3 MΩ. For both Nb and Pb, dJ_n/dJ_{n-1} are identical for $n = 2$ and $n = 3$ and are proportional to $1/R_n \sim T$ as predicted by MAR and MPT.

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[17] Reference [3] gives $dJ_n/dJ_{n-1} = T/16$ for $n > 1$, limiting the ratio to 1/16 for a perfectly transparent barrier. In our experiment we find values larger than this, suggesting that a redefinition of their T is necessary. It seems that the factor 16 is due to an incomplete normalization of the Bogoliubov-Valatin operators. Further, weakly n-dependent factors result from the choice of a specular barrier, which is not appropriate for the present work.

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