Subgap structure in asymmetric superconducting tunnel junctions

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Conductance measurements between two superconducting electrodes with different gap energies Δ1 and Δ2 were performed with a low-temperature scanning tunneling microscope. The temperature dependence and tip-sample distance dependence of the spectra show a pronounced subgap structure which is interpreted as multiple Andreev reflections (MARs). Low temperature conductance peaks not seen in symmetric superconducting tunnel junctions arise at energies ±|Δ1−Δ2| when the superconducting gaps are sufficiently different. We propose an explanation of these findings by extending the full counting statistics of MARs to describe these asymmetric junctions.

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For more than 40 years the subgap structure of superconducting-insulating-superconducting (SIS) junctions has been in the focus of experimental and theoretical investigations.1,2 In these systems the transport is dominated by multiple Andreev reflections.3 These tunneling processes have emerged as a key concept in superconductivity mainly due to recent break-junction experiments.4–7 In these experiments the coupling between the superconductors is varied continuously, a necessity for a thorough understanding of Andreev reflections8,9 and Josephson supercurrent.10

Asymmetric junctions are an unexplored regime in the current framework of Andreev reflections. Because the degeneracy of the gap energies is broken, these systems are predicted to show new spectroscopic features due to the loss and the opening of new Andreev reflection processes.11 Experimental studies of Andreev reflections in asymmetric tunnel junctions would therefore provide crucial insight into subgap processes in SIS systems.

It is possible to study asymmetric tunnel junctions with a scanning tunneling microscope (STM) because the tip and sample can have different gap energies.12 This contrasts with current break-junction techniques, in which the gap energies must be equal. The coupling between the superconductors in STM junctions can be varied by changing the tip-sample distance.13,14 So far, however, Andreev reflections have been detected only in symmetric STM junctions.15

Here we present an experimental study of the subgap structure of asymmetric SIS tunnel junctions. We use a procedure to create the asymmetric SIS junction in an STM by attaching a superconducting microcrystal to a normal conducting tip. Due to finite size effects,16,17 the energy gap of the microcrystal is reduced with respect to the bulk sample value. This technique is particularly powerful because it enables us to vary the asymmetry of the junction by simply using different microcrystals.

In asymmetric niobium junctions created with this technique, we find peaks in the conductance spectra at energies of ±|Δ1−Δ2| at low temperatures. These new features arise at low junction resistances when the difference between Δ1 and Δ2 is sufficiently large. This surprising spectral feature is explained by the extension of the framework of Andreev scattering and the concept of full counting statistics18 to include the asymmetry of the gap energies. The applicability of the MAR model in the asymmetric regime lends strong support to its validity.

The measurements were performed using an STM under UHV conditions in a 3He Joule-Thomson refrigerator.19 We used a Nb(110) single crystal sample which was cleaned by successive cycles of heating and Ar-ion sputtering until we resolved a flat surface.20 The superconducting gap of the sample Δ1=1.47 meV was probed using a cut Ir wire as normal conducting tip. By indenting the Ir tip into the Nb sample we attached a Nb microcrystal on the apex of the tip resulting in a superconducting gap Δ2 ranging from 21–86 % of the bulk Nb value. Using these tips and the Nb(110) sample we performed conductance (dI/dV) measurements by modulating the junction voltage with a small sinusoidal voltage Vmod=10 μVr.m.s and detecting the signal with standard lock-in technique.

Figure 1 shows a set of dI/dV measurements using a tip with a superconducting gap of Δ2=1.27 meV at T=0.56 K and different values of the nominal junction resistance R which were determined outside the gap at V=5 mV. By successively reducing the resistance and thereby increasing the coupling between the two superconductors, we observe the evolution of subgap conductance peaks at characteristic energies of ±Δ1, ±Δ2, and ±(Δ1+Δ2)/3. Also observed is the peak at V=0 corresponding to the Josephson supercurrent. These are observations of MAR in asymmetric superconducting tunnel junctions, which are predicted to occur at energies ±Δ1/m, ±Δ2/m, and ±(Δ1+Δ2)/(2m+1), where m is an integer.11 Processes involving one and two Andreev reflections representing the observed subgap peaks are shown schematically in Fig. 4(a)(i–iii).

The subgap structure observed in Fig. 2 for a tip with a gap Δ2=0.32 meV≈0.21×Δ1 is substantially different. While the peaks at ±Δ1 and ±Δ2 are still present, the peaks at ±(Δ1+Δ2)/3 are only faintly visible. Surprisingly, peaks at ±(Δ1−Δ2), corresponding to ±1.15 meV, appear at lower junction resistances. These features are not present in junc-
The transmission coefficient of states for the tip. As seen in Fig. 3, MAR in asymmetric junctions can account for the observed peak structure, including the feature at $\Delta_1 - \Delta_2$. This is well above the critical temperature of $T_C = 2.1$ K expected for a BCS superconductor with $\Delta = 0.32$ meV. Figure 3(d) shows the unusual behavior of $\Delta_2(T)$, which is presumably due to the small size of the microcrystal. Here $\Delta_2$ was obtained by fitting $dI/dV$ spectra of the tips, measured against a Cu(111) sample, by assuming a bulk BCS density of states for the tip. As seen in Fig. 3(c), such fits reproduce the gap structure of the spectra very well, but have noticeable differences in the tails of the line shape.

To interpret the observed peak structure, we apply a version of the single-channel MAR theory of Ref. 9. We have calculated the conductance for various temperatures and junction resistances by using bulk BCS density of states for the sample and tip with the measured gaps $\Delta_1$ and $\Delta_2$. As seen in Fig. 2, MAR in asymmetric junctions can account for the observed peak structure, including the feature at $\Delta_1 - \Delta_2$.
sum of the individual contributions of the different MAR energies where peaks in the spectrum occur, except at
and the occurrence of tunneling process involving the transfer of
and the annihilation in the left SC. This case is only
neling processes at
Two-reflection process at
1+\Delta_1/3\) involving the tunneling of 3 particles and the creation of a Cooper pair in the right
and the annihilation in the left SC. (iv) Special case of the two-reflection process shown in (iii), where the left Andreev reflection takes place just inside the gap of the left SC. This case is only possible in junctions with \(\Delta_2/\Delta_1\leq 0.5\.

(c) Calculated current contributions \(I_n\) to the overall tunneling current \(I_{\text{sum}}\) (full line) of tunneling processes at \(n\)th order in a junction with \(\Delta_2/\Delta_1=0.21\) and a transmission coefficient of \(\tau=0.2\) at a temperature of 0 K using a full counting statistics calculation. The arrows and the vertical lines mark energies where peaks in the spectrum occur, except at \((\Delta_1+\Delta_2)/3\) (see text).

Similar agreement is obtained for calculations of the temperature dependence shown in Fig. 3(b) by using the measured \(\Delta_2(T)\). Better agreement between the calculated lineshapes and those observed experimentally might be obtained by using a model of the tip density of states that accounts for the differences seen in Fig. 3(c). We note that using more than one conduction channel in the calculation did not improve the agreement of the fits.

To learn which MAR processes contribute to the peak at \(\Delta_1-\Delta_2\), we use the concept of full counting statistics. As shown in Ref. 18, the total current \(I_{\text{sum}}\) can be written as a sum of the individual contributions of the different MAR processes \(I_{\text{sum}}=\Sigma_n I_n\). Here, \(I_n\) is the current contribution of a tunneling process involving the transfer of \(n\) electron charges and the occurrence of \(n-1\) Andreev reflections, and it can be expressed as

\[
I_n(V)=\frac{2e}{\hbar} \int_0^{E_F} n P_n(V,E)dE,
\]

where \(P_n(V,E)\) is the probability of the \(n\)-order tunneling process. The MAR probabilities can be obtained by means of a generalization to the asymmetric case of the recipe described in Ref. 18. In Fig. 4(c) we plot the total current and the main contributions \(I_n\) for a junction with a gap ratio \(\Delta_2/\Delta_1=0.21\). We assume zero temperature and a transmission coefficient of \(\tau=0.2\), which corresponds to a junction resistance \(R=64.5 \text{ k}\Omega\). The conductance peak at \(\Delta_1-\Delta_2\) originates mainly from the large increase of \(I_3\) at this voltage. As illustrated in Fig. 4(b)(iv), such an increase is due to the fact that for \(eV>\Delta_1-\Delta_2\) the two Andreev reflections involved in this MAR process can occur inside the gaps, which implies an enhancement of their probability. Fourth-order processes also contribute strongly to the feature at \(\Delta_1-\Delta_2\); the peak in \(I_4\) evident in Fig. 4(c) results in a marked change in the line shape of the \(dI/dV\) peak at \(\Delta_1-\Delta_2\). So, in short, we propose that the peak at \(\Delta_1-\Delta_2\) is due to the enhancement of the probability of a MAR that transfers three electron charges and involves two Andreev reflections.

On the other hand, in contrast to the data shown in Fig. 1, where the ratio between the gaps is nearly equal, none of the \(I_n\) shown in Fig. 4(c) produces a significant feature at \((\Delta_1+\Delta_2)/3\). The jump in \(I_3\) is only about \(10^{-2} \times I_{\text{sum}}\) because at the onset of this process one of the two Andreev reflections takes place outside the gap, which makes this process quite unlikely [Fig. 4(b)(iii)].

Figure 5 presents in a color coded map the calculated Andreev reflections (\(dI/dV\) maxima) for a SIS tunnel junc-
tion of transmission coefficient $\tau=0.25$ as a function of normalized junction voltage for gap ratios $\Delta_2/\Delta_1$ between 0.05–0.95. At energies of $\Delta_1$, $\Delta_2$, and $\Delta_1+\Delta_2$, Andreev reflections are developed at all ratios. For a ratio $\geq 0.3$ a maximum at $(\Delta_1+\Delta_2)/3$ is clearly detectable, while it diminishes for smaller ratios. A maximum at $\Delta_1-\Delta_2$ occurs for all $\Delta_2/\Delta_1 \approx 0.5$ and vanishes completely when the gap ratio exceeds 0.5. This calculation is in excellent agreement with our observed spectral features presented in Figs. 1 and 2. For the former with a ratio of $\Delta_2/\Delta_1=0.86$ three peaks inside the gap are located at $\Delta_1$, $\Delta_2$, and $(\Delta_1+\Delta_2)/3$, while the peak at $\Delta_1-\Delta_2$ does not exist. For the latter, we observe the $\Delta_1-\Delta_2$ peak, while the peak at $(\Delta_1+\Delta_2)/3$ is only faintly visible.

To summarize, using a low-temperature STM for the creation and characterization of asymmetric superconducting tunnel junctions we gain new insight into the physics of Andreev reflections by analyzing in detail the observed subgap structure. Especially, for junctions with a relatively small gap ratio $\Delta_2/\Delta_1$ we observe peaks at $eV=\pm(\Delta_1-\Delta_2)$, which are not due to the thermal excitation of quasiparticles. All observed subgap features can be understood as Andreev reflections within a full counting statistics model.

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23. If quasiparticle tunneling were the mechanism responsible for the peak at $\Delta_1-\Delta_2$ at low temperatures, then the peak would not diminish at high $R$, as it does in Fig. 2, and it would appear independently of the gap ratio $\Delta_1/\Delta_2$.
24. Line shapes do not include the supercurrent peak because we include only dissipative current in the calculation. For Figs. 2 and 3 we have used a small imaginary part of the energy ($0.01\Delta_1$) to simulate the voltage modulation of the experiment.