Observation of Multiple Andreev Reflections in Superconducting Tunnel Junctions

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(Received 28 October 1993)

We describe measurements on high-current-density Nb-AlO$_x$-Nb tunnel junctions and demonstrate that the development of excess subgap current with increasing barrier transparency is the result of multiple Andreev reflections. We present a model for the barrier consistent with measured junction resistance, the high transparency required for multiple traversals, and the observed single-particle and Josephson characteristics. We argue that excess subgap current and subharmonic gap structure in superconducting tunnel junctions are generally likely to be due to pinhole defects in the barriers.

PACS numbers: 74.50.+r, 73.40.Gk, 73.40.Jn, 85.25.Cp

Increasing the critical current density J_c of Josephson tunnel junctions compensates for their high intrinsic capacitance, allowing device size and response times to be reduced and resulting in improved performance. We recently demonstrated [1] Nb-AlO$_x$-Nb junctions with J_c as high as 4 mA/μm^2, extending the state of the art by an order of magnitude. These devices exhibited significant subgap current. Such "leakage" has been of interest for decades [2,3]. It has long been associated with subharmonic gap structure (SGS), features in the current-voltage (I-V) characteristics at voltages $2\Delta/ne$, where Δ is the superconducting energy gap and n is an integer. Although a variety of mechanisms have been postulated, only three processes consistent with SGS have been seriously touted as explanations for the observed systematic deterioration of tunneling characteristics with increasing J_c: Josephson self-coupling (JSC) [4,5], multiparticle tunneling (MPT) [6], and multiple Andreev reflection (MAR) [7,8].

In nontunneling weak links, MAR is generally accepted as the cause of SGS [7,8]. Although experimental and theoretical characteristics always differ significantly, agreement has been achieved for the *locations* of SGS features in weak links [9], for $n \leq 6$. MAR in weak links is usually treated using a model due to Klapwijk, Blonder, and Tinkham (KBT) [7,8], who considered superconducting junctions with normal interlayers and barriers at the superconductor-normal interfaces. The relevance of this model to tunnel junctions is questionable, however. The normal layer results in a deficit current [10] at large voltages for low values of barrier transmittance [8], and neither the normal layer nor the deficit current occurs in tunnel junctions.

Although reports of SGS in tunnel junctions have existed for years [3], its origin remains controversial [11-13]. SGS is typically observed only for $n \leq 3$, precluding detailed comparisons with theory. JSC can be ruled out in most cases because the predicted resonant current peaks [5] are absent and because the magnetic field dependence is not consistent with Josephson currents. MPT is frequently invoked to explain excess subgap currents, e.g., in recent studies of Nb-AlO$_x$-Nb junctions [11,12]. Although Arnold [14] has shown theoretically that MAR can cause SGS in tunnel junctions, it has been claimed that MPT can better account for experimental observations [12]. However, in order to provide sufficient fitting parameters, MPT-based models make the *ad hoc* assumption that regions of the junction differing not only in barrier thickness, but also in energy gap, contribute to the total current. Furthermore, a recent study [13] showed that the emergence of SGS is accompanied by excess currents at large voltages. Such currents can result from MAR, but not from MPT.

In this paper, we show that MAR is responsible for the excess subgap current in Nb-AlO$_x$-Nb tunnel junctions. We demonstrate excellent agreement with theory for SGS up to large values of n, and propose a model for the barrier which accounts for the near-unity barrier transmittance required for MAR, yet is consistent with the relatively high measured normal state resistance. It accounts for several important features in the I-V characteristics, including the lack of any J_c dependence in their shape over a wide range of J_c (0.2-4 mA/μm^2) and the near-ideal magnetic field dependence of the critical current. We believe this to be the first clear demonstration of MAR in tunnel junctions, and know of no prior work on any junction type showing comparable agreement between experimental and theoretical characteristics.

Junctions with J_c in the range 1 μA/μm^2-4 mA/μm^2 were fabricated by a process described earlier [1]. As J_c increased beyond several tens of μA/μm^2, the I-V characteristics consistently exhibited increasing deviations from the near-ideal behavior of low-J_c devices, including SGS,

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excess subgap current, and excess current at large voltages. The SGS was almost identical in hundreds of junctions fabricated by several processes, with trilayers grown in four different deposition systems under a variety of conditions (e.g., varying Nb stress, process temperature, etc.).

Figure 1 shows the voltage dependence of I and dV/dI at 4.2 K, with I normalized by the Josephson critical current I_c, for representative 1×1 μm^2 junctions with $J_c=0.25$ and 3.1 mA/μm^2. The current rise at $2\Delta/e$ and the SGS at $2\Delta/ne$ occur at lower voltages in the junction with higher J_c. This effect grows with increasing current, and the electrodes of the 3.1 mA/μm^2 junction were driven normal beyond $\simeq 2$ mV. When these characteristics are recalculated with the dependence of Δ on bias due to nonequilibrium quasiparticle injection accounted for [15], the dV/dI curves for the two junctions are virtually indistinguishable when plotted against eV/Δ. Studies of many junctions led to the conclusion that tunneling characteristics, excluding nonequilibrium effects, are independent of J_c over the range $\simeq 0.2$–4 mA/μm^2. For higher J_c's, non-Josephson barrier shorts occurred.

We believe that the subgap current in these junctions is due to MAR, which enables current to flow across a tunnel barrier at voltages $<2\Delta/e$, even at $T=0$. As illustrated in Fig. 2 for the case $n=2$, electrons (holes) originating in one electrode, incident on the other with energy within the superconducting gap, can Andreev reflect as holes (electrons) [16]. Repetition of this process, reversing the roles of the electrodes at each step, results in particles emerging into allowed states in the second electrode. Processes involving n $(n-1)$ Andreev reflections, and $n+1$ (n) total barrier traversals, for even (odd) n result in current increases at voltages $\simeq 2\Delta/ne$. The total current due to the process involving n barrier traversals is reduced by a factor of $(|T|^2)^{n-1}$, where $|T|^2$ is the usual barrier transmittance, with respect to the current due to direct tunneling, so SGS is only significant in junctions in which $|T|^2$ approaches unity. The shape of the conductance-voltage curve provides an "Andreev spectrum" characteristic of a particular barrier transparency. Because the form of the SGS in our junctions does not change for $0.2<J_c<4$ mA/μm^2, the same conduction paths, *with the same* $|T|^2$, dominate the current over this J_c range.

For a tunnel barrier, $|T|^2 \sim \exp(-2Kd)$, where $K=(2mE_B)^{1/2}/\hbar$, E_B is the barrier height, d is the barrier thickness, and m is the carrier mass, provided $2Kd \gg 1$. This condition is met by even a single monolayer of Al oxide, since $E_B \simeq 1.5$ eV (Ref. [17]) and $d \simeq 0.35$ nm. Thus $|T|^2$ is small for any oxide thickness. Because MAR requires that $|T|^2 \rightarrow 1$, we propose that the barrier consists of pinhole defects, metal-metal point contacts (PC) with high $|T|^2$, in parallel with oxide barrier regions having $|T|^2 \ll 1$. Only the PC's contribute to SGS. Because $|T|^2$ decreases rapidly with oxide thickness, PC's contribute significant conductance, especially at subgap voltages, when they cover more than a fraction of 1% of the junction area. In order to model a device, only two parameters are required: $|T|^2$ for the defects and the ratio of the total contribution to the junction conductance from PC and barrier regions.

Most discussions of tunneling through insulators make

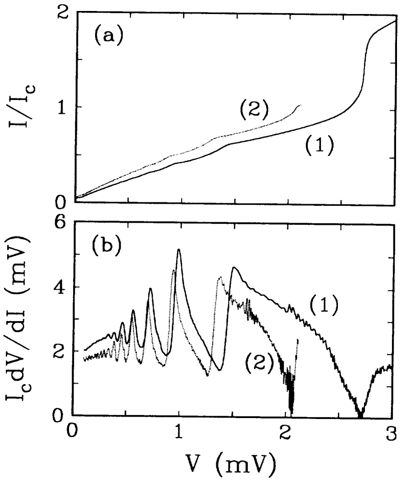

FIG. 1. (a) I-V and (b) dV/dI-V characteristics of (1) 0.25 and (2) 3.1 mA/μm^2 junctions, 1×1 μm^2 (I_c is suppressed with a magnetic field and I is scaled by the original I_c). Only part of curve (2) is shown because the electrodes were driven normal beyond 2 mV. Except for nonequilibrium effects, the characteristics of these junctions are essentially identical.

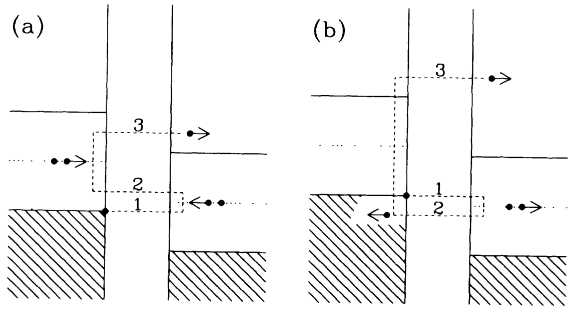

FIG. 2. Schematic of the process involving two Andreev reflections and three barrier traversals (after Ref. [14]). (a) $eV<\Delta$: An electron incident from the left has a finite (holelike) amplitude at the right electrode. It can Andreev reflect as an electron, with the destruction of a Cooper pair. The process is repeated at the original electrode. The resulting electron can now propagate in an allowed state to the right. (b) $eV>\Delta$: An electron incident from the left has a finite (electronlike) amplitude at the right electrode. It can Andreev reflect as a hole, creating a Cooper pair on the right. The reflected hole undergoes a partial Andreev reflection because its energy is near the gap edge, resulting in electron current to the right and hole current to the left. The created Cooper pair represents a net increase of 2 charges flowing compared with process (a). Therefore the current increases near $V=2\Delta/2e$.

the unphysical assumption that barrier thickness is a continuously varying parameter. Real barriers, composed of monolayer (or submonolayer) structural units, must be very nonuniform. Locally, the resistance is quantized, with a value determined by the number of monolayers present. Specific tunneling resistance is given by [18]

$$r_n = 4\pi \frac{h}{e^2} \frac{d^2}{1+2Kd} e^{2Kd} . \qquad (1)$$

A 1 or 2 monolayer thick barrier has a resistance of 0.6 or 110 $\Omega\,\mu\mathrm{m}^2$ (and a J_c of 3.4 mA/μm^2 or 19 μA/μm^2, since $I_cR_n = J_cr_n \simeq 2$ mV for Nb-AlO$_x$-Nb junctions). Thus, even in low-J_c junctions, the barriers are less than 3 monolayers thick, and the existence of pinhole defects in high-J_c junctions is not unexpected. The dependence of J_c on oxidation pressure P and time t, $J_c \propto (\mathrm{Pt})^{-0.4}$, reported in Ref. [1] is due to changes in the local barrier thickness. The much steeper dependence $J_c \propto (Pt)^{-1.6}$ evidently reflects incomplete coverage of the bare Al surface by oxide in the initial stages of growth, giving rise to the PC contribution to junction current. Of course, at too high a density, the PC's coalesce into larger non-Josephson microshorts.

Figure 3 compares the subgap characteristic of the 0.25 mA/μm^2 junction of Fig. 1 with the predictions of our model, using $|T|^2 = 0.68$ and assuming that the defects contribute 30% of the total normal state conductance ($\Delta = 1.44$ meV, very close to the bulk value for Nb at 4.2 K, was used). The model predicts the subgap current level, the locations and approximate amplitudes of the SGS, the size of the step at the gap, and the amount of excess current. The lack of agreement in the vicinity of the Nb gap voltage is due to the proximity effect and nonequilibrium gap reduction. Neither effect is included in the model, but neither is expected to affect this discussion significantly. We also observed elevated I_cR_n values [1], consistent with Arnold's predictions [19].

The characteristics of an unusually "leaky" low-J_c (20 μA/μm^2) junction, with subgap current $\simeq 5$ times larger than usual, are shown in Fig. 4. The dynamic resistance is qualitatively similar to that of Fig. 1(b), even though the level of subgap current is much smaller than that in high-J_c (> 0.2 mA/μm^2) junctions. Also shown are fits to the data using our model, assuming that 4% of the normal state current is due to PC's. The fits are even better than those of Fig. 3; the agreement in the Nb gap region is improved because nonequilibrium effects are negligible. Leaky junctions with lower J_c, and even lower subgap current, show reduced SGS, typically only small features at small values of n.

The conductance of a PC is given by Sharvin's formula [20],

$$r_p = \tfrac{4}{3} \rho l , \qquad (2)$$

where ρ is the metal resistivity and l the elastic mean free path. For Nb [21], $\rho l = 0.31$ m$\Omega\,\mu$m^2 (for $|T|^2 = 1$). We estimated above that $|T|^2 \simeq 0.68$ in our high-J_c devices, so the PC resistance is roughly 0.5 m$\Omega\,\mu$m^2. Thus, for our high-J_c junctions, with J_c and r_n in the ranges 0.2–4 mA/μm^2 and 10–0.5 $\Omega\,\mu$m^2, respectively, transport is dominated by PC's, covering 0.005%–0.1% of the junction area.

We showed earlier [1] that I_c had a nearly ideal (Fraunhofer) dependence on magnetic field, even for 1 × 1 μm^2 areas. This requires that a minimum of several hundred PC's be distributed over a junction area [15], placing an upper limit of less than 1 nm, or roughly one unit

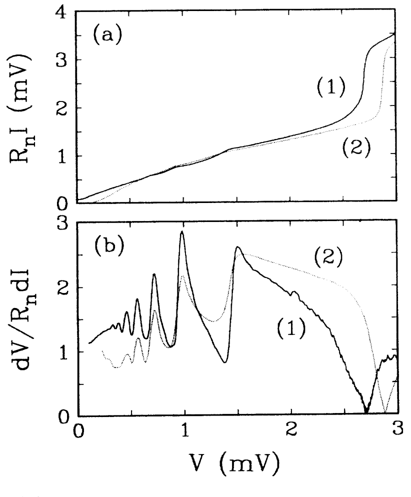

FIG. 3. (a) I-V and (b) dV/dI-V characteristics of a 0.25 mA/μm^2 junction. Curves (1) are experimental data; curves (2) are our model, assuming 30% of the normal state current to be due to PC's.

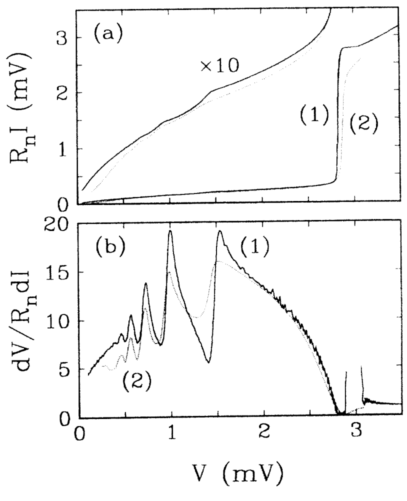

FIG. 4. (a) I-V and (b) dV/dI-V characteristics of a "leaky" 5×5 μm^2, 20 μA/μm^2 junction, with I_c suppressed with a magnetic field. Curves (1) are experimental data; curves (2) are our model, assuming 4% of the normal state current to be due to PC's.

cell, on the PC size (assuming hundreds of PC's in a 1×1 μm^2, 0.2 $mA/\mu m^2$ junction). This, and the fact that the same characteristics are observed under a wide variety of conditions, suggests that the PC's are not random holes in the oxide, but instead are naturally occurring, *reproducible* barrier defects. These structures should be observable by direct imaging of an as-grown barrier by techniques such as atomic force microscopy or ballistic electron emission spectroscopy. In addition, as junction size is reduced, the number of defects should fall until near-ideal Josephson behavior no longer occurs. This should occur first in relatively low-J_c, ~ 0.2 $mA/\mu m^2$, junctions, due to the small defect density. Finally, the dependence of supercurrent on phase difference should become nonsinusoidal [19] at low temperatures, leading to deviations from the Fraunhofer pattern, changes in Shapiro steps under microwave exposure, and deviations from the Ambegoakar-Baratoff [22] temperature dependence of I_c.

In conclusion, the agreement between our model and experiment on the shape of the subgap dynamic resistance and the existence of both excess current and large $I_c R_n$ products, neither of which are consistent with MPT nor JSC, clearly demonstrates that the subgap "leakage" in our junctions results from MAR. We propose that unit-cell-sized defects in the barrier oxide are responsible, and that such structures can account for observed deviations from ideality in all types of tunnel junctions. Barrier pinholes in junctions exhibiting SGS have been suggested before [7], but no barrier model compatible with observed junction properties has been advanced. In our model, unlike MPT-based ones, the junction characteristics are determined only by the transmittance of these regions and the fraction of the junction area they cover (the thinnest part of the oxide contributes the rest of the current); the electrode energy gap is assumed to be constant over the junction area. Elimination of pinhole-type defects in high-J_c junctions should allow reduction of subgap currents, and a monolayer barrier thickness should result in J_c in the several $mA/\mu m^2$ range (note, however, that the increased hysteresis resulting from low subgap currents would actually be detrimental in many Josephson applications).

The authors acknowledge useful conversations with M. Bhushan, K. A. Delin, D. K. Lathrop, and K. K. Likharev and the assistance of K. Stawiasz. This work was conducted under the auspices of and with partial support from the Consortium for Superconducting Electronics, which is partially supported by the Defense Advanced Research Projects Agency (Contract No. MDA 972-90-C-0021).

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