Extreme Critical-Temperature Enhancement of Al by Tunneling in $Nb/AlO_x/Al/AlO_x/Nb$ Tunnel Junctions

M. G. Blamire, E. C. G. Kirk, and J. E. Evetts

Department of Materials Science and Metallurgy, University of Cambridge, Pembroke Street, Cambridge CB2 3QZ, United Kingdom

T. M. Klapwijk

Department of Applied Physics, University of Groningen, Nijenborgh 18, 9747 AG Groningen, The Netherlands

(Received 19 March 1990)

High-conductance $Nb/AlO_x/Al/AlO_x/Nb$ tunnel-junction devices with the Al thickness varying from 3 to 20 nm have been fabricated using a whole-wafer processing route. Current-voltage measurements show a step in the subgap structure corresponding to the difference in the energy gaps $V=2(\Delta_{Nb}-\Delta_{Al})$ of the two superconductors. The measured Δ_{Al} is large and persists to temperatures considerably in excess of the equilibrium critical temperature of the Al. This is interpreted as arising from a tunnel-induced gap enhancement which is an order of magnitude larger than has been seen previously.

PACS numbers: 74.50.+r

The enhancement of superconductivity by both microwave irradiation[1-3] and tunnel injection[4-7] has been the subject of considerable attention. Although large gap enhancements under tunnel injection have been predicted,[8-11] existing experimental results have shown enhancements of less than 5% of the equilibrium value at any temperature.

In the experiments reported in this paper we have detected the presence of a strongly superconducting state under subgap quasiparticle injection at $V=2(\Delta_{Nb}-\Delta_{Al})$ and $T \leq 4.2$ K in *S-I-S′-I-S* devices (*S*, superconductor; *I*, insulator) whose Al S' layer varies between 3 and 20 nm, and has a maximum equilibrium critical temperature ($T_{c\mathrm{eq}}$) less than 2.4 K. This effect has been observed in all devices of this type. $T_{c\mathrm{eq}}$ is measured by determining the highest temperature at which the zero-voltage supercurrent is present. We have shown that the maximum Δ_{Al} in this state corresponds to the value which would be measured at $T=0$ and that it does not follow the equilibrium BCS temperature dependence. In contrast, at zero voltage the temperature dependence of the critical current agrees with the equilibrium description provided by Eq. (1) below. We have further shown that the temperature dependence of the current at the step corresponding to $V=2(\Delta_{Nb}-\Delta_{Al})$ fits with Eq. (2) below and thus represents an enhanced gap state. We are thus observing effective T_c enhancements of approximately 100%, over an order of magnitude larger than values previously reported.[6] In this Letter we will describe the experimental details and results, discuss the measurement of $T_{c\mathrm{eq}}$, and provide a model for the behavior observed.

For our experiments we have used results obtained from $Nb/Al/AlO_x/Al/AlO_x/Al/Nb$ structures deposited as a sequence without breaking vacuum. Multilayer films containing the two tunnel barriers were deposited on *R*-plane sapphire substrates in a dual-target UHV sputtering system described elsewhere.[12] The tunnel barriers were formed by thermally oxidizing the lower two Al layers to a specific resistance of $(3\text{–}7)\times10^{-11}$ $\Omega\,\mathrm{m}^2$ with a maximum conductance ratio of 3:1. An angled shield partially screened the 12.5-mm × 3-mm chips from the Al magnetron, producing a variation of Al thickness along the substrate length in all three Al layers. Seven mesas were patterned at equal 1.5-mm spacings along each of the substrates and a CF_4 barrel etch and anodization process was performed to isolate both tunnel barriers in each device.[13] This allowed devices with different S' thicknesses to be prepared and tested as a single operation. For comparison, previous experiments have used soft alloy structures with tunnel conductances ranging from as low as 1×10^{-9} Ω m (Refs. 2, 4, and 7) to 4×10^{-10} Ω m (Refs. 2 and 6) and typical S' layer thicknesses have been of the order of 30 nm. In addition, by using two similar or equal conductance barriers and superconductors with very different equilibrium gaps we have been able to more closely approach the symmetrical quasiparticle extraction conditions envisaged by Parmenter[8] in his original concept.

Current-voltage measurements were performed on each double-barrier device using a 70-Hz ac current bias circuit. The Al thickness between the barriers was measured by precision anodization spectroscopy[14] on large areas exposed by CF_4 etching close to each device. Magnetic fields of up to 10 mT could be applied parallel to the plane of the tunnel barriers and the devices could be cycled in liquid He between 2.2 and 4.2 K in a storage Dewar using a lambda-plate insert.

At 4.2 K and for sufficiently thick (≥ 10 nm) Al layers, the devices showed a characteristic resembling two *S-I-N* (*N*, normal metal) junctions of similar, but nonidentical, conductances in series [see Fig. 1(a)]—no subgap structure is observed and there is no evidence of a supercurrent even under high magnification. The similarity of the conductances allows the two junctions to be simultaneously biased on the steep current rise at $V=\Delta_{\rm Nb}/e$. However, even at 4.2 K, for thicknesses <10 nm, the shape of the I-V plot alters and shows a sharp gap structure and a pronounced step in the subgap [shown in Fig. 1(b)]. The voltage at which this step occurs increases as the Al thickness increases, and is temperature dependent. As the temperature is reduced towards 2.2 K, the I-V characteristics of devices with Al thicknesses between 10 and 20 nm increase in sharpness and present a similar subgap peak [also shown in Fig. 1(b)]. At still lower temperatures the I-V plot in 3–10-nm-Al devices resolves into a double S-I-S' characteristic, with a symmetrical Fraunhofer dependence of the zero-voltage critical current (I_c) on magnetic field indicating that complex flux-trapping effects are not responsible for its nonappearance at higher temperature; this is shown in Fig. 1(c).

Below $T_{c\rm eq}$ the double S-I-S' characteristic appears to be perfectly normal, with a subgap peak (appearing as a step under current bias) at the difference of the two gaps $V=2(\Delta_{\rm Nb}-\Delta_{\rm Al})$. In our devices the barrier conductances are sufficiently similar for there to be no evidence of a second peak at $V=2\Delta_{\rm Nb}-\Delta_{\rm Al}$. It is this subgap step structure which persists to higher temperatures above $T_{c\rm eq}$, and which provides the evidence for the persistence of a tunneling-induced superconducting state. The very small temperature dependence of this subgap step suggests that in this biased condition the superconducting state in the Al is very strongly enhanced by quasiparticle tunneling [up to $\Delta_{\rm Al}(0)$]. This paper will discuss the observed effect in terms of the existing theory of gap enhancement, and show that the conditions for large enhancement are satisfied in our devices.

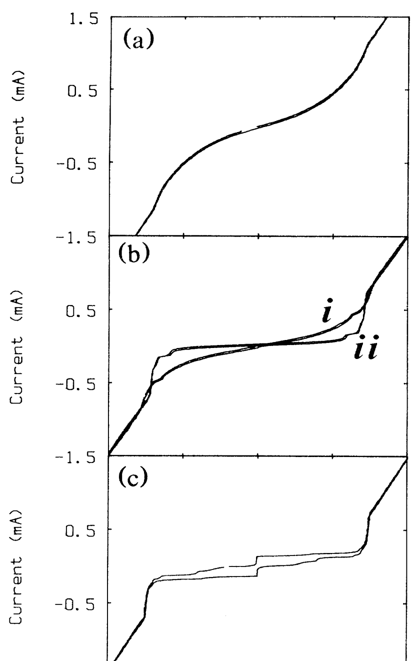

FIG. 1. Electrical response of 28-μm^2 Nb/AlO$_x$/Al/AlO$_x$/Nb devices. (a) 12-nm Al layer at 4.2 K; (b) curve i, 3-nm Al layer at 4.2 K; curve ii, 12-nm Al layer at 2.2 K; (c) 7-nm Al layer at 2.2 K. The tunnel barriers had conductances in the ratio 3:1, resulting in the apparently nonasymptotic normal resistance.

The correct measurement of the equilibrium superconducting transition temperature ($T_{c\rm eq}$) of the central Al layer is of crucial importance to these results. This was carefully measured by noting the temperature at which a zero-voltage supercurrent appeared; in this state quasiparticle T_c-enhancement effects are absent and the temperature dependence of the critical current below T_c should follow the Ambegaokar and Baratoff expression[15]

$$I_c=R_n^{-1}\Delta' K([1-\Delta_{\rm Al}(T/T_c')^2/\Delta_{\rm Nb}(T/T_c)^2]^{1/2})\,, \qquad (1)$$

where R_n is the normal-state tunneling resistance. A temperature dependence of this type was observed, implying that this is a valid measure of $T_{c\rm eq}$, and in all cases $T_{c\rm eq}<2.4$ K. While proximity effects should be possible across tunnel barriers and may result in a modified T_c under zero-voltage current bias, such effects will (through similarity to a conventional S-N-S junction) result in the presence of a critical current at a temperature ***higher*** than the true equilibrium critical temperature and thus our measurement (if affected in this way) will be an *overestimate* of $T_{c\rm eq}$. Moreover, if proximity effects were important, then above the true $T_{c\rm eq}$ a temperature dependence more closely resembling that of an S-N-S device would be expected; this is not observed.

We are confident therefore that the devices we have fabricated consist of a low-equilibrium-T_c Al layer sandwiched between two tunnel barriers. The external electrodes are Nb/Al proximity structures and because of the geometry of the system the Al thickness of these layers varies in a similar manner to the central layer; it was never larger than 7 nm. However, Huggins and Gurvitch[16] report only minor changes in Nb/Al/AlO$_x$/Nb device characteristics even with Al proximity layers as large as 50 nm; thus the effects reported in this paper originate as a result of changes in the middle Al S' layer, and the external electrodes may be treated as behaving as pure Nb.

The presence of the feature $V_{\rm step}=2(\Delta_{\rm Nb}-\Delta_{\rm Al})$ at temperatures as high as 4.2 K indicates that these structures are very far from equilibrium. In Fig. 2 we show the values of the gap measured by the position of this step as a function of temperature for the four devices shown in Table I; the measurement error arises largely as a result of imprecision in locating the top left edge of the step. This shows that $\Delta_{\rm Al}$, although varying from device to device, is approximately constant at temperatures

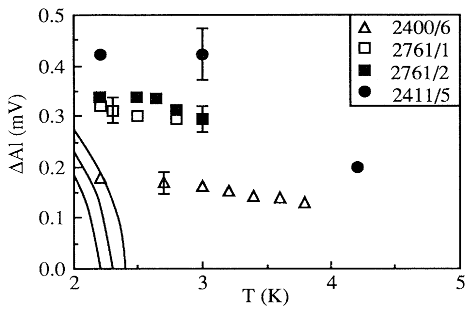

FIG. 2. Plot of the Al energy gap as a function of temperature for the same devices shown in Table I. Sample error bars are shown for each data set. For comparison the solid lines show the BCS equilibrium-temperature dependence of the gap corresponding to $T_{c\mathrm{eq}}$ values of (from left) 2.2, 2.3, and 2.4 K.

up to 3 K and then decreases progressively. That this is not a manifestation of the equilibrium properties of the Al is shown by the data in Table I: Here the maximum value of Δ_{Al} (taken at 2.2 K) is shown together with T_{ceq} for the Al. In addition, we can use the data to calculate values for $2\Delta_{\mathrm{Al}}/kT_{c\mathrm{eq}}$ for the four different devices; these values are shown in Table I.

In all cases, the values are of order 3.5, in accordance with the BCS prediction for $2\Delta_{\mathrm{Al}}(0)/kT_c$. Thus we can say that the maximum Δ_{Al} in each case corresponds to $\Delta_{\mathrm{Al}}(0)$, confirming that we are *correctly measuring* $T_{c\mathrm{eq}}$. However, the temperature dependence of this gap near T_c is grossly different from the BCS prediction of $(1-T/T_c)^{1/2}$ (also shown in Fig. 2), implying that the gap continues to be enhanced to a value close to $\Delta_{\mathrm{Al}}(0)$ at temperatures well above $T_{c\mathrm{eq}}$.

The behavior of the devices is fitted by a model in which all thermal electronic excitations in the Al are removed by tunnel extraction, thus maintaining the enhanced superconducting state. Consider two identical *S-I-N* junctions in series. When the tunnel conductance is high, the distribution of quasiparticles in the Al film cannot remain thermal or the forward tunnel conduction of quasiparticles from the Al would greatly exceed the rate of quasiparticle creation via thermal phonons. The spontaneous formation of an energy gap and decrease in

TABLE I. Data for sample devices: $T_{c\mathrm{eq}}$ was the temperature at which the zero-voltage critical current vanished; Δ_{Al}max was the largest measured energy gap for each device.

Device	Al thickness (nm)	$T_{c\mathrm{eq}}$ (K)	Δ_{Al}max (meV)	$\frac{2\Delta_{\mathrm{Al}}\mathrm{max}}{kT_{c\mathrm{eq}}}$
2400/6	20	< 2.2	0.18 ± 0.02	
2411/5	7.3	2.4 ± 0.1	0.42 ± 0.05	4.0 ± 0.5
2761/2	7	2.3 ± 0.1	0.34 ± 0.03	3.4 ± 0.3
2761/1	8	2.2 ± 0.1	0.31 ± 0.03	3.3 ± 0.3

the effective temperature of the Al quasiparticles greatly reduces the number of quasiparticles and thereby decreases the forward tunnel conduction rate to a point where it equals the quasiparticle creation rate. The largest gap and lowest Al temperatures are obtained when the junctions are biased at $\Delta_{\mathrm{Nb}}-\Delta_{\mathrm{Al}}$ because this voltage corresponds to the highest rate of extraction of quasiparticles from the Al to the Nb films. The temperature of the phonons is unchanged or perhaps increases slightly (see below).

At any value of current therefore, a dynamic equilibrium exists between the rate of quasiparticle activation and recombination in the Al layer and the forward quasiparticle tunnel current. The maximum possible tunnel current for any particular value of Δ_{Al} thus occurs when the activation and tunnel rates are equal, and both occur at a higher rate than recombination.

In our system an equality of this type is expected to occur at $V=2(\Delta_{\mathrm{Nb}}-\Delta_{\mathrm{Al}})$ and $\Delta_{\mathrm{Al}}\approx\Delta_{\mathrm{Al}}(0)$, since the forward tunnel current is high and the Al is superconducting. If we equate the current I_{step} at the top left corner of the step on the positive curve with the rate of quasiparticle excitation, then this rate must be proportional to the density of thermal phonons of the appropriate energy $[\exp(-2\Delta_{\mathrm{Al}}/kT_{\mathrm{ph}}]$, and thus to a good approximation

$$I_{\mathrm{step}}/I_{2\Delta}=Ae^{-\Delta_{\mathrm{Al}}/kT_{\mathrm{ph}}}. \qquad (2)$$

The analysis assumes the conductance of the two barriers to be equal. In fact, the gap enhancement is observed when the conductances differ by a factor of up to 3. This is not surprising, since quasiparticles will be efficiently extracted from the Al layer when the lower conductance barrier is biased at $V=\Delta_{\mathrm{Nb}}-\Delta_{\mathrm{Al}}$. Provided that the conductance is high enough, this will be sufficient to maintain and create a finite gap and result in the formation of a subgap step. The presence of the vertical edge of the step will allow both barriers of different conductances to be simultaneously at $V=\Delta_{\mathrm{Nb}}-\Delta_{\mathrm{Al}}$ while current biased. Although the unequal conductances will result in temporary charge imbalance, once the voltages across the two barriers are equalized the tunnel rate is largely controlled by the quasiparticle excitation rate rather than by the tunnel conductance and the imbalance will mostly relax. Clearly, the greater the ratio of conductances the smaller the range of current over which this condition can be maintained, but a factor of 3 does not seem extreme in this context.

A major uncertainty in any experiment of this type is the possibility of phonon trapping in the S' layers. The simpler models assume that thermally excited phonons escape immediately from S', whereas previous experiments have detected significant trapping effects in which the excess of phonons serve to "reheat" the S' layer. However, as noted by Gray[17] there is evidence that the importance of trapping decreases as the Al film thickness

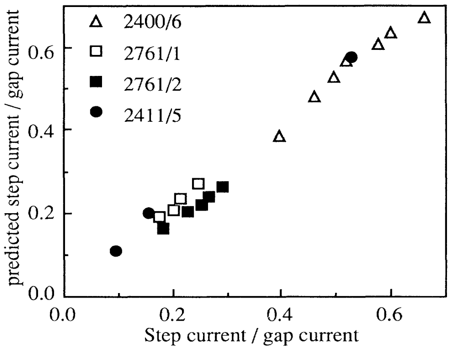

FIG. 3. The measured current at the top of the subgap step normalized with respect to the current rise at $2\Delta_{Nb}$ plotted against the value predicted by Eq. (2) using the measured gap at each temperature. Device data shown in Table I. The errors bars have been removed for clarity, but are of the order of the symbol size horizontally, and twice the symbol size vertically.

is reduced. Evidence that the trapping effect must be small in our devices is provided by the linearity obtained by the use of the equilibrium temperature in calculating the data in Fig. 3; trapping would raise the effective temperature above this value.

We have presented clear experimental evidence for large gap-enhancement effects in our devices. The Nb/Al system is ideally suited to such studies since Al has one of the largest values of τ_0, and the difference in the equilibrium gaps is large; it should, however, be possible to observe it using other materials.

Funding for part of this work was provided by the European Space Agency (Contract No. 7898/88/NL/PB). We would like to thank Dr. R. E. Somekh, Dr. J. H. Claassen, and Dr. G. W. Morris for careful reading of the manuscript. M.G.B. is supported by the Royal Society, and E.C.G.K. by the Interdisciplinary Research Center (IRC) in Superconductivity, Cambridge.

[1]T. M. Klapwijk and J. E. Mooij, Physica (Amsterdam) **81B**, 132 (1976).

[2]J. J. Chang and D. J. Scalapino, J. Low Temp. Phys. **29**, 477 (1977).

[3]J. E. Mooij, N. Lambert, and T. M. Klapwijk, Solid State Commun. **36**, 585 (1980).

[4]K. E. Gray and H. W. Willemsen, J. Low Temp. Phys. **31**, 911 (1978).

[5]C. C. Chi and J. Clarke, Phys. Rev. B **19**, 4495 (1979).

[6]K. V. Mitsen, Fiz. Tverd. Tela (Leningrad) **21**, 3475 (1979) [Sov. Phys. Solid State **21**, 2007 (1979)].

[7]I. Iguchi and D. N. Langenberg, Phys. Rev. Lett. **44**, 486 (1980).

[8]R. H. Parmenter, Phys. Rev. Lett. **11**, 274 (1961).

[9]A. G. Aronov and V. L. Gurevich, Zh. Eksp. Teor. Fiz. **36**, 1809 (1973) [Sov. Phys. JETP **36**, 957 (1973)].

[10]V. F. Elesin and Yu. N. Kopaev, Usp. Fiz. Nauk **133**, 259 (1981) [Sov. Phys. Usp. **24**, 116 (1981)].

[11]G. M. Eliashberg and B. I. Ivlev, in *Non-equilibrium Superconductivity*, edited by D. N. Langenberg and A. I. Larkin (North-Holland, Amsterdam, 1985).

[12]M. G. Blamire, R. E. Somekh, H. Barber, G. W. Morris, and J. E. Evetts, J. Appl. Phys. **64**, 6396 (1988).

[13]M. G. Blamire, R. E. Somekh, and J. E. Evetts, IEEE Trans. Magn. **25**, 1135 (1989).

[14]M. G. Blamire, K. H. Huang, R. E. Somekh, E. C. G. Kirk, G. W. Morris, and J. E. Evetts, Appl. Phys. Lett. **55**, 732 (1989).

[15]V. Ambegaokar and A. Baratoff, Phys. Rev. Lett. **10**, 486 (1963).

[16]H. A. Huggins and M. Gurvitch, J. Appl. Phys. **57**, 2103 (1985).

[17]K. E. Gray, Appl. Phys. Lett. **32**, 392 (1978).