Observation of a novel, inelastic, charge-imbalance relaxation process in superconductor–insulator–normal-metal tunnel junctions

Thomas R. Lemberger and Yeouchung Yen
Physics Department, Ohio State University, Columbus, Ohio 43210
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A recently predicted inelastic charge-imbalance relaxation process associated with the proximity of a normal metal to a superconductor is confirmed through measurements of the low-voltage dc resistance of superconductor–insulator–normal-metal tunnel junctions. The process is quantitatively linked to the pair-breaking nature of the proximity effect through the measured depression in T_c caused by the normal electrode.

In the past dozen years, a great deal of theoretical and experimental work on the nonequilibrium phenomenon of charge imbalance in superconductors has revealed the importance of charge imbalance in the transport of an electric current across the surface of a superconductor.[1–15] One crucial parameter in determining the relative importance of charge imbalance in a particular situation is the charge-imbalance relaxation rate. The magnitude and temperature dependence of this rate is determined by the various electron scattering processes that may be present, for example, inelastic electron-phonon scattering and elastic exchange scattering from magnetic impurities. Because tunnel junctions have been used in many studies of charge-imbalance phenomena, it is very important to understand how the presence of a counterelectrode affects charge-imbalance relaxation in the superconducting electrode.

A recent theory[14,15] predicts that, contrary to earlier assumptions, the N electrode in a superconductor–insulator–normal-metal (SIN) tunnel junction does contribute to the total charge-imbalance relaxation rate. In fact, if the coupling between the S and N electrodes is strong enough, then this proximity-effect relaxation process can be much more important than bulk scattering processes. A key feature of the theory is the prediction that the magnitude and temperature dependence of the low-voltage dc resistance of a SIN junction is directly related to the depression in the transition temperature caused by the normal electrode. In this Rapid Communication, measurements on three very-low-resistance Al-AlO_x-Cu junctions are presented and compared with the results of a numerical calculation based on the theory.

The sample configuration is illustrated in Fig. 1. The shape of each film was defined with an aperture mask. The Al (200 Å) film was evaporated onto a clean, room-temperature glass substrate. Its narrow section was about 2 mm long and 320 μm wide. Silicon monoxide (500 Å) defined the junction width of about 320 μm. The normal counterelectrode was 2000–3000 Å thick, and it was made by evaporating a mixture of roughly 98% Cu, 1% Al, and 1% Fe to completion from a W boat. The Fe was necessary to prevent a supercurrent from flowing from the Al film into a Pb (1000 Å) overlayer on the counterelectrode. From visual observations of the color of the counterelectrode during its deposition, we believe that the Fe evaporated after the Cu, as expected from the respective vapor pressures of Fe and Cu at the evaportion temperature of about 1000 °C. The circles in Fig. 1 represent a 6-mm-diam Pb (2000 Å) ground plane centered on the junction and isolated from the sample by a 2000-Å-thick SiO film. The film thicknesses were determined from a thickness monitor that uses a vibrating quartz-crystal sensor.

The sample was immersed in liquid ^{4}He. A superconducting quantum interference device (SQUID) was used in a nulling feedback mode to measure the voltage across the junction (Fig. 1). Mercury batteries were used for current sources. The current-voltage (I-V) characteristics of the junctions were plotted on an XY recorder, with typical bias voltages of less than 50 nV. Below this voltage, the I-V characteristics were linear to within the measurement sensitivity of 1%, except when the bias current exceeded the critical current I_c of the Al in the junction. The junction resistance $R_j(T)$ was determined from the slope of the I-V curve, and I_c was defined to be the bias current at which the I-V curve broke sharply from linearity.

Data for sample 1 are shown in Fig. 2. Because $R_j(T)$ diverges as $1/(T_c-T)^{1/2}$ for T near T_c, it is convenient to plot the square of the conductance of the junction $G_j\equiv 1/R_j$. Then G_j^2 extrapolates linearly to zero at a temperature that we define to be the transition temperature T_c of the Al in the junction. This definition of T_c is supported by the observation that the critical current I_c, plotted as open circles in Fig. 2, also goes to zero at T_c. We determined the critical current I_{c0} of the Al not in the junction from the current at which the I-V characteristic of the strip switched from zero to a finite voltage. The transition temperature T_{c0} of the Al not in the junction was determined by extrapolating $I_{c0}(T)$ to zero by eye. I_{c0} is plotted as solid circles in Fig. 2. The dashed curves in Fig. 2 are the theoretical maximum values for I_c and I_{c0}, calculated by using measured parameters and by assuming a uniform

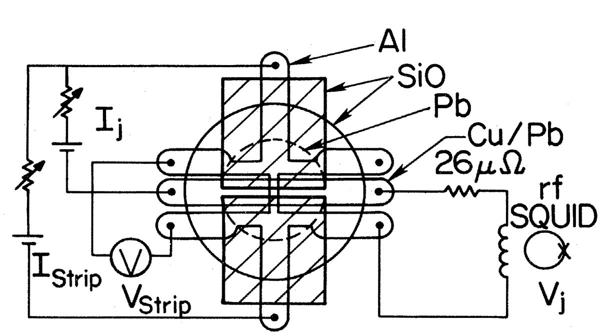

FIG. 1. Sample geometry and measurement configuration.

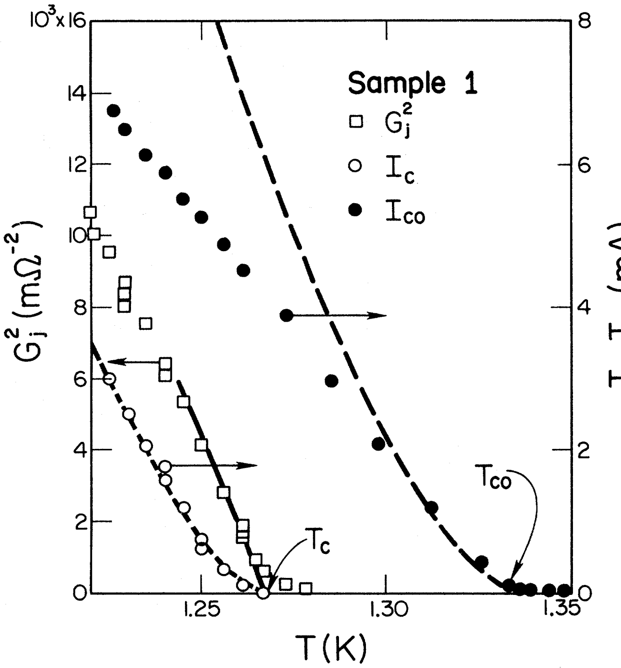

FIG. 2. Measured values of the square of the junction conductance G_j^2 (□); the critical current of the Al in the junction I_c (○); and the critical current of the Al not in the junction I_{c0} (●). The solid line shows how T_c is defined from G_j^2, and the dashed lines are the maximum theoretical values of I_c and I_{c0}.

current density.[16] These curves show that the measured critical current is that of the bulk of the film.

The parameters for sample 1 (2) (3) were as follows: electron mean free path[16] $l = 4\times10^{-16}\ \Omega\,\mathrm{m}^2/\rho(4.2\ \mathrm{K}) = 13.0$ nm (10.5 nm) (10.3 nm); film thickness $d = 20$ nm (20 nm) (20 nm); $T_c = 1.267$ K (1.387 K) (1.321 K); $T_{c0} = 1.333$ K (1.400 K) (1.387 K); $R_i(T_c) = 3.45\ \mu\Omega$ (9.00 $\mu\Omega$) (3.52 $\mu\Omega$), determined from G_j as described below. Finally, the inelastic electron-phonon scattering rate for electrons with the Fermi energy at $T = T_c$ was estimated to be

$$1/\tau_E \approx (T_{c0}/1.2\ \mathrm{K})^3/12\ \mathrm{ns} = 1.1\ (1.3)\ (1.3)\times10^8/\mathrm{s}\ .$$

The theory of the low-voltage resistance of SIN tunnel junctions is based on the Boltzmann equation approach of Pethick and Smith.[4] Near T_c, the physical picture is especially clear. The bias current generates an electric potential drop in the insulator and it generates a nonequilibrium state characterized by a charge imbalance in the S electrode. The presence of a charge imbalance implies an excess of quasiparticles on one side of the Fermi surface and a deficit on the other side. Therefore, there is a nonzero density of quasiparticle charge. To maintain local charge neutrality, the density of the condensate charge must change by the opposite amount by adjusting its chemical potential. This results in a chemical potential drop across the junction, in addition to the electric potential drop.

The resistance of the junction is the sum of two parts, corresponding to the electric and the chemical parts of the electrochemical potential: the resistance of the insulator $R_i(T)$, that approaches a finite value linearly as $T\rightarrow T_c$; and a resistance $R_{Q^*}(T)$ associated with the charge imbalance, that diverges as $1/(T_c-T)^{1/2}$ as $T\rightarrow T_c$. The divergence in R_{Q^*} can be understood physically by imagining the bias current to be fixed at some small value while the temperature is increased towards T_c. The size of the charge imbalance, and also the chemical potential drop, generated by the current is proportional to the charge-imbalance relaxation time τ_{Q^*}. As $T\rightarrow T_c$, τ_{Q^*} continuously approaches its normal-state value, which is infinite because a charge imbalance can neither exist nor relax in the normal state. Hence, the measured voltage diverges.

To compare our results with theory, we numerically solved the Boltzmann equation describing a SIN junction biased at a small voltage V,[15] including electron-phonon scattering and the nonequilibrium parts of the quasiparticle injection term. The total electric current was calculated from the resulting quasiparticle distribution function by using Eq. (2.15) of Pethick and Smith.[4] There are two important physical parameters in the theory. One is the ratio $\tau_E/\tau_{\mathrm{tun}}$ of the characteristic rate associated with quasiparticle tunneling,

$$1/\tau_{\mathrm{tun}} = 1/2N(0)e^2\Omega R_i(T_c)\ , \tag{1}$$

to the electron-phonon scattering rate $1/\tau_E$. In Eq. (1), $2N(0)$ is the volume density of electron states in the S electrode, e the electron charge, Ω the area A of the junction times the thickness d of the superconducting electrode, and $R_i(T_c)$ the resistance of the insulator at $T = T_c$. For the samples reported here, $\tau_E/\tau_{\mathrm{tun}} >> 1$, so that the tunneling relaxation rate is very important.

The other important parameter is the ratio $\delta T_c/T_{c0}$, where δT_c is the depression in the transition temperature due to pair breaking. $\delta T_c/T_{c0}$ is related to the total pair-breaking rate $1/2\tau_E + 1/2\tau_{\mathrm{tun}}$ through[15]

$$\delta T_c/T_{c0} = (\pi\hbar/4k_BT_{c0})(1/2\tau_E + 1/2\tau_{\mathrm{tun}})\ . \tag{2}$$

If this ratio is larger than about 0.2, then pair-breaking effects on the superconducting density of states and order parameter must be included in the calculation. For the three samples reported here, $\delta T_c/T_{c0} < 0.05$, so that the BCS density of states and order parameter were used in the calculation.

The end result of the calculation is the normalized junction conductance,

$$g_j(t) = R_i(T_c)/R_j(t)\ , \tag{3}$$

as a function of the normalized temperature $t \equiv T/T_c$. Near $t = 1$, the calculated values of g_j agree with the analytic result:[14]

$$g_j^2 = 1/(1 + 4k_BT/\pi\Delta)^2 \sim 5.8(1-t)\quad (t \gtrsim 0.98)\ . \tag{4}$$

A graph of g_j^2 in the limit $\tau_E/\tau_{\mathrm{tun}} >> 1$ is shown as a dashed curve in Fig. 3; the solid line is discussed below. Experimental values of g_j^2 are also shown in Fig. 3, where $R_i(T_c)$ is a fitting parameter determined from the slope of g_j^2 near T_c by using Eq. (4). Clearly, the measured values of $g_j(T)$ have the predicted temperature dependence for $T \gtrsim 0.96T_c$, but they deviate from theory at lower temperatures.

Before discussing the deviation, we emphasize that it is very important to check the theory quantitatively by comparing the measured value of $\delta T_c/T_{c0}$ with the value calculated from Eqs. (1) and (2) by using measured sample parameters. From the experimental values for T_{c0}, Ω, $R_i(T_c)$, and $1/\tau_E$ listed above, and the literature value[17]

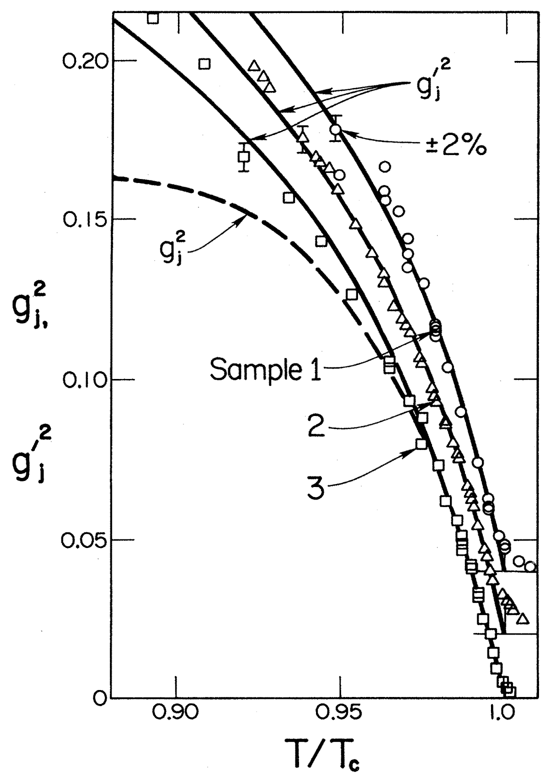

FIG. 3. Measured values of the square of the normalized junction conductance of samples 1(○), 2(Δ), and 3(□). The dashed curve represents g_j^2, the square of the normalized junction conductance calculated numerically. The solid line represents $g_j'^2$, a modified conductance discussed in the text. Note that the data and the theory curves for samples 1 and 2 are displaced vertically by 0.04 and 0.02 units, respectively, for clarity.

$2N(0)=3.48\times10^{28}/\mathrm{eV\,m}^3$, for sample 1 (2) (3) we find $\delta T_c/T_{c0}=0.056$ (0.020) (0.051). These values agree very well with the directly measured values 0.050 (0.009) (0.048). The measurements near T_c, therefore, confirm quantitatively the existence of an inelastic charge-imbalance relaxation process associated with the proximity of a normal metal to a superconductor.

The solid lines in Fig. 3 represent a modified junction conductance:

$$g_j'(t)=R_i(T_c)/[R_j(t)-R_i(t)+R_i(T_c)] \ . \qquad (5)$$

In effect, g_j' differs from g_j in that the temperature-dependent BCS[18,19] resistance of the insulator $R_i(t)$ has been replaced by its value at T_c, $R_i(T_c)$, in the calculated value of the total junction resistance, $R_j(t)$. We have no explanation for why g_j' fits the data better than g_j. We can note only that measurements[20] on junctions with a specific resistance $[R_i(T_c)A]$ 300 times larger than those presented here also were fitted better by assuming that R_i was independent of T.

In conclusion, data on three SIN tunnel junctions clearly demonstrate an inelastic charge-imbalance relaxation process associated with the proximity of a normal metal to a superconductor. Furthermore, they quantitatively link this process to the pair-breaking nature of the proximity effect. This process may be important in any measurement of inelastic scattering rates involving superconductors in proximity to normal metal.

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