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Observation of phase fluctuations and electron-electron scattering in weakly disordered superconducting aluminum films

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Phase fluctuations and electron-electron scattering in weakly disordered, two-dimensional superconducting Al films have been observed through their effects on the resistance of low-resistance Al-AlO_x-Cu tunnel junctions. These are the uniform-film analog of the fluctuations in percolating films and in Josephson junctions. Disorder-enhanced electron-electron scattering has the magnitude and temperature dependence expected theoretically.

We present measurements of the resistance R_j of very low resistance Al-AlO_x-Cu, i.e., superconductor-insulator-normal metal (SIN), tunnel junctions in which the Al film is weakly disordered. R_j is presented as a function of temperature T and at fixed T, as a function of the extrinsic pair-breaking rate $1/\tau_s$ generated by an applied supercurrent and by supercurrents induced by a magnetic field.

These measurements probe the pair-breaking effects of disorder on uniformly thick, homogeneous superconducting films in the superconducting state, i.e., below T_c and B_{c2}. Pair breaking refers to processes which broaden the peak in the electron density of states and cause other deviations from Bardeen-Cooper-Schrieffer (BCS) theory,[1,2] e.g., elastic exchange scattering from magnetic impurities and inelastic electron-phonon scattering. These measurements are complementary to studies of the effects of disorder on the phase boundary in the B-T plane[3-8] which involve non-pair-breaking effects as well. They cannot be compared easily with earlier, conventional tunneling measurements on high-resistance junctions with disordered but less uniform films: granular Al films[9] and extremely thin Sn (Ref. 10) and Pb films.[11]

We have two major new results. First, we observe a new pair-breaking mechanism from supercurrents associated with thermal fluctuations in the phase of the order parameter. Second, we observe for the first time the magnitude and strong temperature dependence of the electron-electron scattering rate below T_c, in agreement with theory,[12,13] and measurements on normal[14-16] Al films, and in disagreement with values from superconducting[17] Al films near T_c, taken with a different technique.

Our novel tunneling technique has been described in detail.[18,19] The idea is that R_j for low-resistance SIN junctions contains a significant "nonequilibrium" portion $R_{Q^*}(T)$ associated with the quasiparticle charge imbalance generated in the S film by the measuring current. $R_{Q^*}(T)$ reflects the magnitude and T dependence of all pair-breaking processes, and can be probed precisely by applying an extrinsic pair breaker such as a supercurrent.

The theory[18] of charge imbalance in SIN junctions has been verified in detail by measurements on Sn and SnIn films,[20] which serve as a simple model systems. Measurements of the effect of supercurrents on high-resistance SIN junctions,[21] in which R_{Q^*} is negligible, also are in excellent agreement with theory and demonstrate our understanding of pair breaking caused by applied supercurrents.

The configuration of our SIN junctions is illustrated in Fig. 1. Junctions were built on thick anodized Nb films (on glass substrates) to increase the measured critical current of the Al films. The electron mean free path l of the Al films was shortened by evaporation at ≈ 4 Å/s in about 3×10^{-5} Torr O_2. The Al films were oxidized in about 5×10^{-4} Torr O_2 for about a minute immediately after deposition. Cu films were evaporated with Au (3 wt.%) and Fe (1 wt.%) to block the diffusion of Cooper pairs from the Pb to the Al film. The Pb film on the Cu made the current through the junction as uniform as possible.

$R_j(T)$ was determined from the slope of the linear current-voltage (I-V) characteristic. Measurements of R_j as a function of supercurrent I_s in the Al strip and magnetic field $B_{\parallel}$ parallel to the substrate were made by fixing the junction current I and measuring the change in voltage V. V was always less than 100 nV to avoid heating.

Measured sample parameters and experimental results for two samples on which we have the most complete data are shown in Table I. For later reference, note that $l \ll \xi_0$, where $\xi_0 = \hbar v_F/\pi\Delta_0 \approx 1600$ nm is the pure-limit coherence length, so dirty-limit expressions for the Ginzburg-Landau coherence length $\xi(T)$, and the penetration depth $\lambda(T)$, are appropriate. The supercurrent density should be uniform through the thickness d of the Al film because $2\lambda(T) \gg d$. Because the thermal coherence length

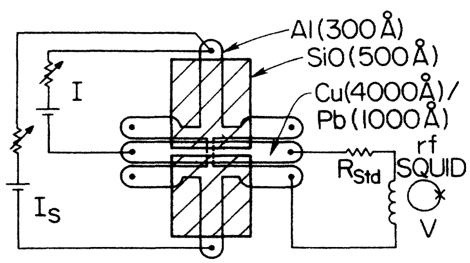

FIG. 1. Sample configuration. I and V are the current and voltage in the junction, I_s is the current in the Al film. Junction area is 300×300 μm^2. The Nb ground plane is not shown.

TABLE I. Sample parameters and experimental results. $R_\square$ is the resistance per square of the Al film at 4.2 K, l is calculated from $\rho l = 4\times10^{-16}$ $\Omega\,\mathrm{m}^2$ (Ref. 38). Values of times and rates are all at T_c. $1/\tau_{e\text{-}e}^{\mathrm{expt}}(T_c)$ is the fitted value of the electron-electron scattering rate; $1/\tau_{e\text{-}e}^{\mathrm{th}}(T_c)$ is calculated from the definition [Eq. (2)]. $1/\tau_{\mathrm{SCF}}^{\mathrm{expt}}(T_c)$ is the fitted value; $1/\tau_{\mathrm{SCF}}^{\mathrm{th}}(T_c)$ is calculated from Eq. (3).

Sample	$R_\square$ (Ω)	d (Å)	T_c (K)	l (Å)	$\tau_{e\text{-}e}^{\mathrm{th}}(T_c)/\tau_{e\text{-}e}^{\mathrm{expt}}(T_c)$	$\tau_{\mathrm{SCF}}^{\mathrm{th}}(T_c)/\tau_{\mathrm{SCF}}^{\mathrm{expt}}(T_c)$
SH13	2.4	343	1.480	49	1.7	0.9
SH17	2.3	422	1.519	41	1.0	1.2

$(D\hbar/k_BT)^{1/2} > d$, electron-electron scattering in the Al film is two dimensional.[14,15,22]

Figures 2 and 3 illustrate the excellent agreement between model calculations and our data. Figure 2 shows $R_j(T)/R_n$ vs T/T_c for sample SH13 fitted with a curve calculated with the scattering rates (at T_c) shown. Figure 3 shows R_j at several representative temperatures as a function of the square root of the extrinsic pair-breaking rate $1/\tau_s$ caused by an applied supercurrent I_s and magnetic field $B_\parallel$. These (additive) pair breakers can be expressed as[18]

$$1/\tau_s = \frac{2\gamma^2}{\pi^4}\frac{R_\square}{2N(0)(k_BT_c)^2dw^2}\frac{n_s(0,0)^2}{n_s(T,1/\tau_s)^2}I_s^2 + \frac{d}{12N(0)\hbar^2R_\square}B_\parallel^2\,, \qquad (1)$$

where n_s is the the density of superconducting electrons (calculated in the dirty limit[1]), w is the width of the Al film, and γ is $\Delta(0)/k_BT_c \approx 1.76$. Note from Fig. 3 that R_j has precisely the same dependence on $1/\tau_s$ for supercurrents and magnetic fields, as expected for uniformly thick, homogeneous films. This agreement confirms that variations in thickness that affect the local current density in the films are unimportant. As a quick guide to Fig. 3, we note that the inflection point occurs when the extrinsic applied pair-breaking rate is about half of the intrinsic rate.[18] This allows an experimental determination of the inelastic electron-electron scattering rate $1/\tau_{e\text{-}e}^{\mathrm{expt}}(T_c)$ and the pair-breaking rate from supercurrent fluctuations $1/\tau_{\mathrm{SCF}}^{\mathrm{expt}}(T_c)$ from the different temperature dependences of these rates below T_c, as found from the model calculations described below.

Although difficult to describe in detail in this limited space, with guidance from the literature concerning the magnitude and energy dependence of the electron-phonon and electron-electron scattering rates, there is sufficient data to determine the sample parameters T_c and R_N, and the rates $1/\tau_{\mathrm{tun}}$, $1/\tau_{e\text{-}e}^{\mathrm{expt}}(T_c)$, and $1/\tau_{\mathrm{SCF}}^{\mathrm{expt}}(T_c)$, with precision. Roughly speaking, the analysis is as follows.

The divergence in $R_j(T)$ reflects the divergence in the charge-imbalance relaxation time expressed through $R_{Q^*}(T)$. It and the critical current of the Al strip give the same T_c to within a few mK. Theory[18] shows that R_N,

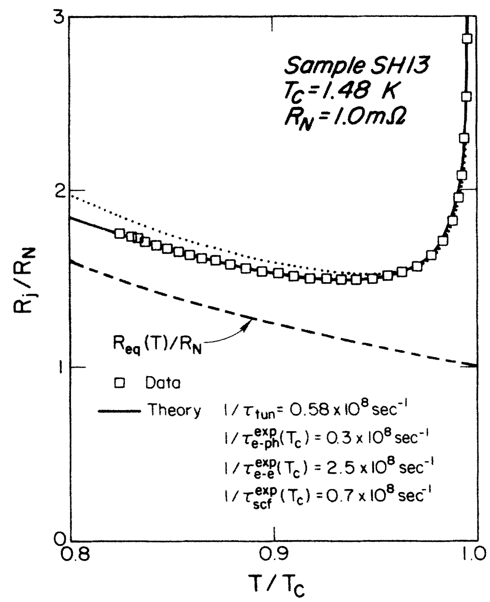

FIG. 2. Normalized junction resistance $R_j(T)/R_N$ as a function of temperature. $R_{\mathrm{eq}}(T)/R_N$ is the normalized junction resistance calculated with quasiparticles in thermal equilibrium. R_N is the intrinsic resistance of the junction. $R_Q^*(T) = R_j(T) - R_{\mathrm{eq}}(T)$. The dotted curve is calculated with $1/\tau_{\mathrm{SCF}}(T_c) = 0$ and $1/\tau_{e\text{-}e}(T_c) = 5.1\times10^8\ \mathrm{sec}^{-1}$, which is 3 times larger than expected from Eq. (2), to show that even an inflated rate cannot explain the data. The other parameters are the same as for the solid curves.

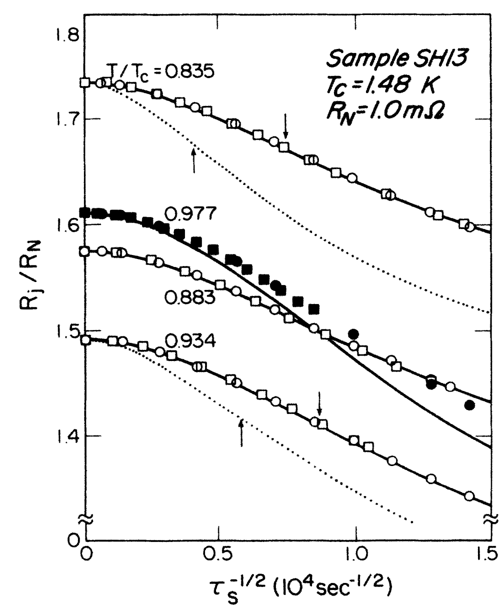

FIG. 3. Junction resistance vs $\sqrt{1/\tau_s}$, where $1/\tau_s$ is the extrinsic pair-breaking rate generated by applied supercurrents (□) and parallel fields (○). $\sqrt{1/\tau_s}$ is proportional to I_s or $B_\parallel$. The solid and dotted curves are calculated with the same parameters as in Fig. 2. The dotted curves are shifted vertically to match the data at $1/\tau_s = 0$. Inflection points are indicated by arrows at two different temperatures.

the intrinsic junction resistance, lies between the minimum value of $R_j(T)$ and half of the minimum value, typically being about 20% below the minimum. R_N determines the tunneling rate $1/\tau_{\rm tun}=1/2N(0)e^2\Omega R_N$, where Ω is the volume of Al in the junction and $2N(0)=3.48\times10^{28}/\mathrm{eV\,m^3}$ is the density of states in Al.[23] Thus, T_c, R_N, and $1/\tau_{\rm tun}$ are determined to about 20% just from $R_j(T)$ near T_c.

Experimental[14–16] and theoretical[24] results on Al films above T_c suggest the value of $1/\tau_{e\text{-ph}}(T_c)$ given in Fig. 2, which is negligible compared with the other rates. We include it for completeness although it has no effect on our calculated curves.

The total intrinsic pair-breaking rate associated with tunneling, electron-electron scattering, and supercurrent fluctuations comes from the value of $1/2\tau_s$ at the inflection point in R_j vs $\sqrt{1/\tau_s}$. Electron-electron and supercurrent fluctuation contributions are separated by fitting R_j vs T and R_j vs $\sqrt{1/\tau_s}$ with model calculations based on a Boltzmann-type equation for the quasiparticle distribution function[18,25] as follows.

First, we consider inelastic scattering processes only. Electron-electron coupling enters through the coupling function (Refs. 16 and 22) $\alpha^2F(\omega)=e^2R_\square/8\pi^2\hbar$ in the scattering integral. From this, we define a characteristic theoretical electron-electron rate for comparison, Table I, with the experimental value:

$$1/\tau_{e\text{-}e}^{\rm th}(T_c)\equiv 7\zeta(3)\frac{e^2R_\square k_BT_c}{2\pi\hbar\hbar}\approx(4.3\times10^7)R_\square T_c[(\Omega\,\mathrm{K\,s})^{-1}]\ . \qquad (2)$$

With this definition, the calculated charge-imbalance relaxation rate near T_c is (Refs. 2 and 26) $1/\tau_{Q^*}=\pi\Delta/4k_BT_c\tau_{e\text{-}e}^{\rm th}(T_c)$ when electron-electron scattering is the dominant charge-imbalance relaxation mechanism. The definition Eq. (2) is similar, but not identical, to the phase-breaking rate involved in electron localization.[16]

The data shown in Figs. 2 and 3 cannot be fitted with only electron-electron scattering. Electron-electron scattering decreases too rapidly below T_c as the gap in the density of states opens.[12] This is illustrated by the dotted curves in Figs. 2 and 3, which were calculated by using an electron-electron rate three times larger than expected form Eq. (2) to give a reasonable fit near T_c. However, at lower T, the data show a larger intrinsic pair-breaking rate than the calculation both in the larger experimental value of $1/2\tau_s$ at the inflection points in Fig. 3 and in the smaller experimental value of $R_{Q^*}(T)$ in Fig. 2.

Next, we include supercurrents as a relaxation mechanism with the same dependence on Δ and quasiparticle energy as for an applied supercurrent,[27] but with a characteristic rate proportional to T. Now the calculations are in excellent agreement with the data (Figs. 2 and 3). As shown in Table I, the fitted value $1/\tau_{e\text{-}e}^{\rm expt}(T_c)$, which is determined primarily from data for $T/T_c\geq0.95$, agrees well with the prediction [Eq. (2)]. The fitted value $1/\tau_{\rm SCF}^{\rm expt}(T_c)$, which comes primarily from $T/T_c\leq0.95$, agrees well with the expression derived below.

We believe that supercurrents associated with thermal phase fluctuations produce pair breaking just as applied supercurrents do. (Several comments[28–30] in the literature support this possibility, but there is no microscopic theory.) We can estimate the pair-breaking rate with the rough approximation that superconducting electrons in volume $\xi^2(T)d$ act as a single particle undergoing Brownian motion. The equipartition theorem gives $m\langle v_s^2\rangle n_s\xi^2d/2\approx k_BT$. From this and the result (Ref. 1) $1/\tau_s=0.5D(2mv_s/\hbar)^2$ for superfluid flow,

$$1/\tau_{\rm SCF}^{\rm th}\equiv\langle 1/\tau_s\rangle\approx\frac{4D}{\hbar^2}\frac{mk_BT}{n_s\xi^2(T)d}\approx(2.2\times10^7)R_\square T[(\Omega\,\mathrm{K\,s})^{-1}]\ , \qquad (3)$$

with dirty-limit expressions for $n_s(T)$ and $\xi(T)$.[31] This agrees well with our fitted value.

An equivalent description involves summing the contributions to $\langle v_s^2\rangle$ from fluctuations with wave vectors k up to $1/\xi(T)$. Microscopically, the fluctuations are driven by fluctuating electric fields and by gradients in charge-imbalance fluctuations.[32] For some values of k and T, they correspond to thermal excitation of Carlson-Goldman modes[33] and propagating plasma modes.[34] For most values of k, the fluctuations are damped. The phase fluctuations are the uniform-film analog of fluctuations in nearly discontinuous percolating superconducting films,[10,35] and in Josephson junctions.[36] They are the precursor of the more violent fluctuations associated with thermal vortex-antivortex pairs that occur in heavily disordered films.[37]

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