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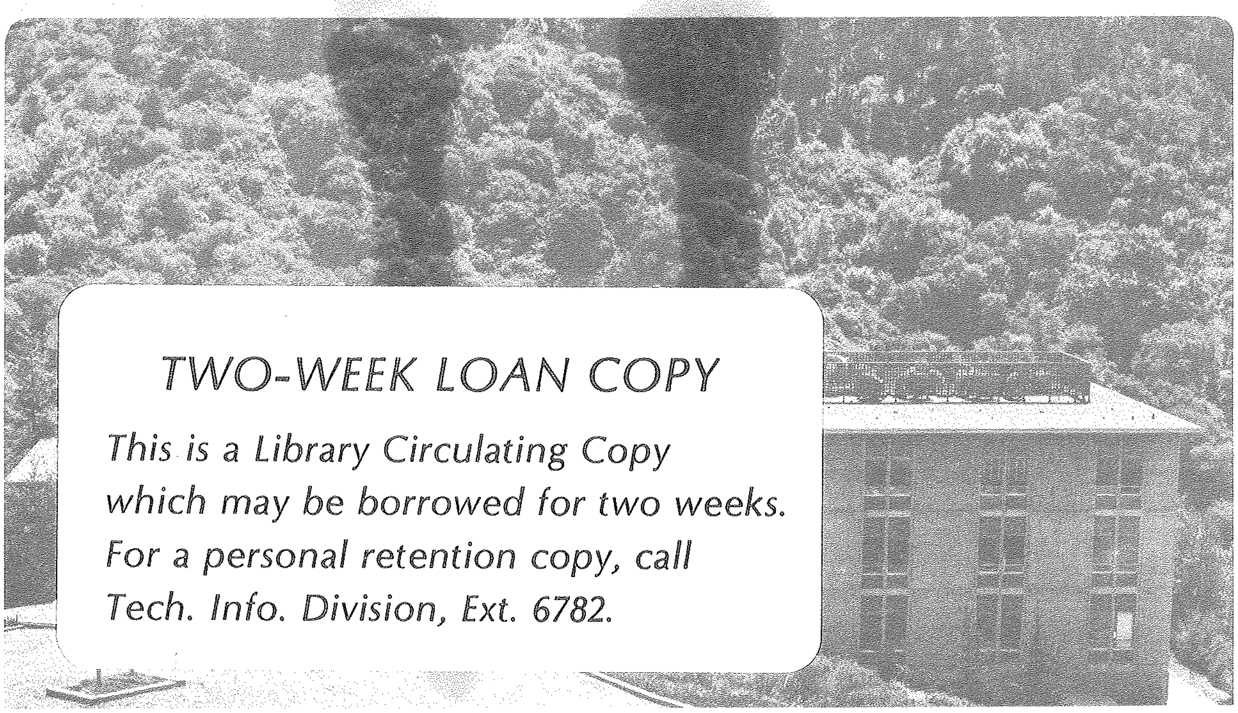

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CHARGE-IMBALANCE FLUCTUATIONS IN SUPERCONDUCTORS

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We show that the mean square amplitude of the fluctuations of the condensate chemical potential, μ_s, due to charge-imbalance fluctuations in the limit $\Delta/k_BT \ll 1$ is $\langle(\delta\mu_s)^2\rangle = 2(k_BT)^2/\pi\Delta\Omega N(0)$ in a volume Ω of superconductor. We relate these fluctuations via Nyquist's theorem to measured values of the contribution of self-injected charge-imbalance to the dc resistance of SIN tunnel junctions to show that the dynamic charge-imbalance relaxation rate is $1/\tau_E$, the electron-phonon scattering rate.

A charge imbalance Q^* exists in a superconductor when the densities of electron-like and hole-like excitations are not equal[1-6]. To maintain charge neutrality when a charge imbalance exists, the chemical potential of the condensate, μ_s, must shift from its equilibrium value, μ_s^0, by an amount[5]

$$\delta\mu_s = -Q^*/2N(0), \qquad (1)$$

where N(0) is the single-spin density of states per unit volume in the normal state. In thermal equilibrium, $\langle \delta\mu_s \rangle = 0$, but $\langle (\delta\mu_s)^2 \rangle \neq 0$. (Angular brackets $\langle\rangle$ indicate a thermodynamic average.) In this Letter, we derive an expression for $\langle (\delta\mu_s)^2 \rangle$ by combining the present theory of charge imbalance[2-6]. with equilibrium thermodynamics. We show that these fluctuations are related via Nyquist's theorem[7] to a contribution to the dc resistance of superconductor-normal metal (SIN) and superconductor-superconductor (SIS) tunnel junctions and to the boundary resistance of superconductor-normal metal (SN) interfaces[8], which arises from the charge imbalance generated in the superconductor by the measuring current. We derive an expression for this extra resistance, which we call R_{Q^*}, appropriate to tunnel junctions. We present measurements of R_{Q^*} which agree with our theory, and use them to determine the bandwidth for charge-imbalance fluctuations.

First we calculate $\langle (\delta\mu_s)^2 \rangle$ in the limit $\Delta/k_BT \ll 1$, where Δ is the order parameter and T is the temperature, and demonstrate that fluctuations in μ_s are independent of those in Δ and T. Since the effects of currents induced by gradients in $\delta\mu_s$ are neglected, the calculation is limited to fluctuations with wavelengths much larger than $\lambda_{Q^*} = (D\tau_{Q^*})^{1/2}$, where $D = \frac{1}{3}v_F\ell$ is the electron diffusion constant and τ_{Q^*} is the static charge-imbalance relaxation time. Thus, our result is not directly applicable to SN interfaces since gradients in $\delta\mu_s$ which are induced in the superconductor by the proximity of the normal metal are neglected.

Consider a volume Ω of superconductor, either an isolated grain or part of a larger system, which has all of its extensive parameters except entropy fixed, and is in contact with a thermal reservoir at temperature T^o. Then equilibrium thermodynamics requires[9]:

$$\langle(\delta\mu_s)^2\rangle = k_B T^o / (\partial^2 F^o/\delta\mu_s^2)\Big|_{T,N,\Delta} \quad , \qquad (2)$$

where $F^o = H - T^o S$ is the appropriate free energy, H and S are the energy and entropy of the electrons in Ω, and the derivative is evaluated in equilibrium.

We evaluate $\partial^2 F^o/\partial\mu_s^2\Big|_{T,N,\Delta}$ as follows. Tinkham[10] derives

$$\hat{H} - \mu_s\hat{N} = \sum_k (\xi_k - E_k + \Delta b_k^*) + \sum_k E_k(\gamma_{k0}^\dagger\gamma_{k0} + \gamma_{k1}^\dagger\gamma_{k1}), \qquad (3)$$

where $\hat{H}$ and $\hat{N}$ are the Hamiltonian and electron number operators, $\xi_k = (\hbar^2 k^2/2m) - \mu_s$, $E_k = +(\xi_k^2 + \Delta^2)^{1/2}$, and $\Delta = V_{BCS}\sum_k b_k$ is assumed to be real and isotropic. Equation (3) can be evaluated at any instant by replacing the quasiparticle operators, $\gamma_{k0}^\dagger\gamma_{k0}$ and $\gamma_{k1}^\dagger\gamma_{k1}$, by f_k, where f_k represents the fraction of quasiparticle states near k which are occupied, and f_k is assumed to be independent of spin and of the direction of k. It is important to remember that the instantaneous values of μ_s, Δ, and T may not be equal to their equilibrium values, μ_s^o, Δ^o, and T^o. Assuming a constant density of states, for simplicity, and weak electron-phonon coupling, we find

$$H - \mu_s N = -N(0)\mu_s^2 - \tfrac{1}{2}N(0)\Delta^2 + N(0)\Delta^2 \ln\frac{\Delta}{\Delta^o(0)} + 2\sum_k E_k f_k, \qquad (4)$$

which is no longer an operator expression. Equation (4) indicates that if each f_k changes by a small amount δf_k, then $H-\mu_s N$ changes by

$$\delta(H - \mu_s N) = -N\delta\mu_s' + 2\sum_k E_k \delta f_k , \qquad (5)$$

where $\delta\mu_s' = \delta Q^*/2N(0)$ is the change induced in μ_s. This implies that for a system with a fixed number of electrons,

$$\delta H = 2\sum_k E_k \delta f_k . \qquad (6)$$

By using the usual expression for the entropy of a system of fermions[9],

$$S = -k_B \sum_k [f_k \ln(f_k) + (1-f_k)\ln(1-f_k)], \qquad (7)$$

along with Eq. (6), we find

$$\delta F^o = \delta H - T^o \delta S = 2\sum_k \left(E_k - k_B T^o \ln \frac{f_k}{1-f_k} \right) \delta f_k . \qquad (8)$$

To make further progress, we must make some approximation regarding the form of f_k. We assume that

$$f_k = 1/ [\exp(E_k'/k_B T) + 1], \qquad (9)$$

and

$$E_k' = \left[\left(\frac{\hbar^2 k^2}{2m} - \mu_n \right)^2 + \Delta'^2 \right]^{\frac{1}{2}}, \qquad (10)$$

where, to lowest order in $\Delta^o/k_B T^o$ [4],

$$\Delta' - \Delta^o = (\Delta - \Delta^o)(1 + 8.4\Delta^o/\pi^3 k_B T^o), \qquad (11)$$

and

$$\delta\mu_n = \mu_n - \mu_s^o = -\delta\bar{\mu}_s \, \pi\Delta^o/4k_B T^o . \qquad (12)$$

Note that our μ_n and $\delta\mu_n$ are identical to those of Pethick and Smith[4]. Equations (11) and (12) are determined from Eqs. (9) and (10) by the gap equation and charge neutrality, respectively. This hydrodynamic approximation should be quite good for small fluctuations in Δ because these should decay in a time much larger than τ_E, where τ_E is the electron-phonon scattering time for a quasiparticle at the fermi energy at $T=T_c$. It is also good for fluctuations in T, provided the thermal response time of the system is much greater than τ_E. The approximation is not strictly applicable for fluctuations in μ_s since, as we show in this Letter, they decay in a time τ_E. Nevertheless, using this approximation in Eq. (8) yields:

$$\delta F^o = 2\sum_k \left(E_k - \frac{T^o}{T} E_k' \right) \delta f_k. \tag{13}$$

Equation (13) indicates that when $\mu_s = \mu_s{}^o$, $\Delta = \Delta^o$, and $T = T^o$, F^o is stationary with respect to changes in f_k, as is required for equilibrium. From Eq. (13) we find

$$\partial^2 F^o/\partial\mu_s\partial\Delta\big|_{T,N} = \partial^2 F^o/\partial\mu_s\partial T\big|_{\Delta,N} = 0, \tag{14}$$

$$\partial^2 F^o/\partial\Delta^2\big|_{T,\mu_s,N} = 4N(0)\Omega(1 - T^o/T_c), \tag{15}$$

and

$$\partial^2 F^o/\partial\mu_s{}^2\big|_{T,N,\Delta} = \pi\Delta^o N(0)\Omega/2k_B T^o. \tag{16}$$

Equation (14) indicates that fluctuations in μ_s are uncorrelated with those in Δ and T. Equation (15) is consistent with the spatically uniform Ginzburg-Landau[11] equations, thus demonstrating the basic physical correctness of the hydrodynamic approximation for f_k. Substituting Eq. (16) into Eq. (2) yields the desired result:

$$\langle(\delta\mu_s)^2\rangle = 2(k_B T^o)^2/\pi N(0)\Omega\Delta^o. \tag{17}$$

In the rest of the paper the superscript which distinguishes equilibrium values from instantaneous values is dropped.

These fluctuations, and the accompanying order parameter phase fluctuations, should be especially important in granular films. For example, for an isolated Al grain with $\Omega = (10\text{nm})^3$, $T_c = 1.5$K, and $\Delta/k_BT = 0.1$, Eq. (17) gives $\langle(\delta\mu_s)^2\rangle^{1/2} = 200\mu\text{eV} \approx 1.5k_BT$. They should also be observable as additional voltage noise across SIN and SIS tunnel junctions. Thus, they are related via Nyquist's theorem to an additional dc resistance, R_{Q*}, which must appear across these devices. We now derive an expression for R_{Q*} and present experimental results which verify our expression.

When a dc current I flows through a junction, the voltage V across the junction has a component, $\delta\mu_s/e$, due to the steady-state charge imbalance $Q^* = F^*\tau_{Q*}/Ie\Omega$, generated in the superconducting electrode by I. By using Eq. (1), we find

$$R_{Q*} = \delta\mu_s/eI = F^*\tau_{Q*}/2N(0)\Omega e^2. \qquad (18)$$

The notation here is the same as that used in Refs. 12, 13, and 14. The quantities F^* and τ_{Q*} depend on T, V, and whether the junctions are SIS or SIN. Note that R_{Q*} is in series with, and independent of, the familiar tunnel oxide resistance, which we call R_{ox} for clarity.

Figure 1 shows measured values of the low-voltage ($|V| \leq 100$nV) dc resistance of two Al(~1300Å)-AlO_x-Cu(7000Å) tunnel junctions versus measured values of Δ/k_BT. The samples were essentially the same three-film, two-tunnel-junction configurations used by Lemberger and Clarke[12,13], with SiO used to define the SIN tunnel junction area to be 0.82x0.82 mm^2. The Al film parameters for sample 1(2) were T_c=1.243K (1.240K), electron mean free path

ℓ=43 nm (43 nm), and film thickness d=127 nm (136 nm). The mean free path was determined from the measured resistivity by using[15] $\rho\ell = 9\times10^{-16}\ \Omega m^2$. As has been observed before[12,16] for low specific resistance SIN tunnel junctions, the low-voltage value of R_{ox} is nearly constant over a wider range of values of Δ/k_BT near $\Delta/k_BT=0$ than expected[17]. We therefore fit the data with $R^{th} = R_{ox} + R_{Q*}$, where R_{ox} is a constant and F^* and τ_{Q*}/τ_E have been calculated for SIN junctions in the limit $|eV| \ll \Delta, k_BT$ by Clarke <u>et al.</u>[18], and Chi and Clarke[14], respectively. Note that there are no adjustable parameters in R_{Q*} since Ω is measured, N(0) is taken from the literature to be[19] 1.74×10^{28}/eV m^3, and τ_E can be calculated from[14] $\tau_E = 12ns(1.2K/T_c)^3$ to be 11 ns for both films. Equation (18) gives $R_{Q*}\tau_E/F^*\tau_{Q*} = 23\ \mu\Omega$ (21 $\mu\Omega$) for sample 1(2). The only adjustable parameter is R_{ox}. As illustrated in Fig. 1, the resulting fit is quite good for $\Delta/k_BT \lesssim 0.5$. (Incidentally, R_{Q*} was observed as a dip in conductance near $T=T_c$ by Clarke and Paterson[16] and by Lemberger and Clarke[12] in their detector junctions. Thus, these authors should have replaced their measured normalized conductances g_{NS} by unity for temperatures at which the dip was observed. However, since the measured dips were small, this correction would have a negligible effect on their results.)

Nyquist's theorem relates the low-voltage value of R_{Q*}, which we measured, to the amplitude of voltage fluctuations across the unbiased junction due to charge-imbalance fluctuations in the volume of superconductor adjacent to the tunnel oxide, which we calculated:

$$\langle (\delta\mu_s/e)^2 \rangle = 4k_BTR_{Q*}B, \qquad (19)$$

where B is the bandwidth for the fluctuations. If an externally generated charge imbalance would decay exponentially in a time τ, then τ is the dynamic charge-imbalance relaxation time, and $B=1/4\tau$. Equation (19) applies to junctions which: 1) have areal dimensions $>>\lambda_{Q*}$ and superconducting film thicknesses $d<<\lambda_{Q*}$; 2) have an RC time constant $<<\tau_E$; 3) have a normal-state resistance $R>>\tau_E/2N(0)e^2\Omega$, which ensures that presence of the junction does not affect the bandwidth of the fluctuations; and 4) are at temperatures sufficiently below T_c that the superfluid response time[6] $\tau_o<<\tau_E$. Our junctions satisfy these criteria for $\Delta/k_BT \gtrsim 0.05$.

We can determine B as follows. Having verified Eq. (18), we can obtain an analytic expression for R_{Q*} by approximating the numerically calculated values of F^* and τ_{Q*} with the analytic forms[3] valid for $\Delta/k_BT<<1$, $\tau_{Q*} = 4k_BT\tau_E/\pi\Delta$ and $F^* = 1$, to obtain $R_{Q*} \approx 4k_BT\tau_E/2N(0)\Omega e^2\pi\Delta$. This result and Eq. (17) can be substituted into Eq. (19) to yield $B=1/4\tau_E$. Therefore, the dynamic charge-imbalance relaxation time is τ_E, and not the steady-state charge-imbalance relaxation time τ_{Q*}. This surprising result was predicted by Kadin et al.[6], and this is the first supporting experimental evidence.

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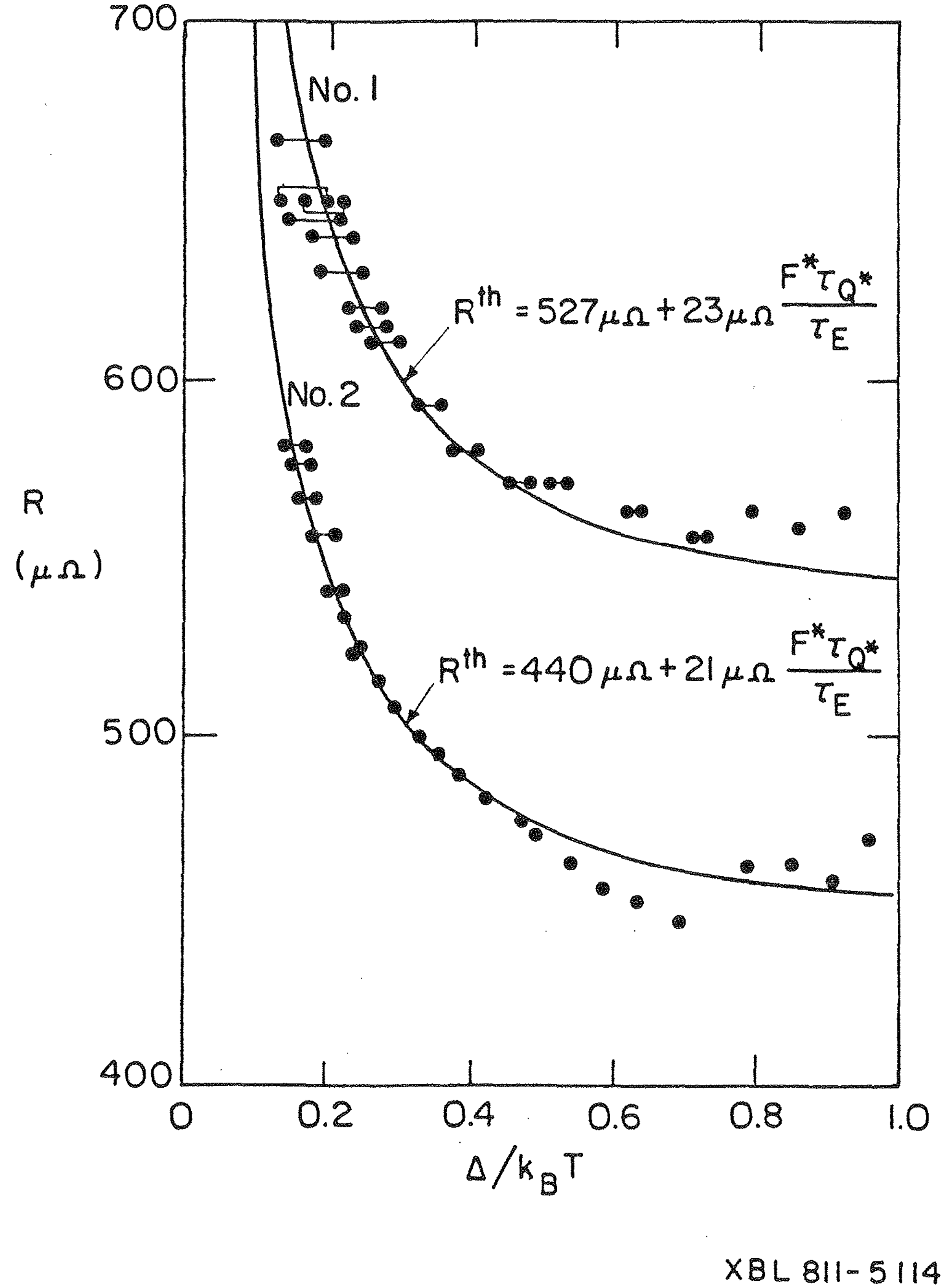
FIGURE CAPTION

Fig. 1 Plot of junction resistance R vs. Δ/k_BT measured for two Al-AlO$_x$-Cu tunnel junctions. The solid curves represent a theoretical fit to the data. Note the suppressed zero on the vertical axis.



700
600
500
400
R
(μΩ)
No. 1
No. 2
R^th = 527μΩ + 23μΩ F*τ_Q*/τ_E
R^th = 440 μΩ + 21 μΩ F*τ_Q*/τ_E
0
0.2
0.4
0.6
0.8
1.0
Δ/k_B T
XBL 811-5114

Fig. 1