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CHARGE-IMBALANCE RELAXATION IN THE PRESENCE OF A
PAIR-BREAKING SUPERCURRENT IN DIRTY, SUPERCONDUCTING
AL FILMS.

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ABSTRACT

The charge-imbalance relaxation rate, $1/F^*\tau_{Q^*}$, has been measured in
dirty Al films in the presence of an applied supercurrent for $0.7 \leq T/T_c \leq 0.91$. The
supercurrent $I_s$ induces an anisotropic energy gap, and hence allows non-spin-
flip elastic scattering to relax the charge imbalance. The effect of a super-
current is equivalent to that of magnetic impurities, with a pair-breaking
rate $\tau_s^{-1} = D(p_s/h)^2/2$, where $D$ is the electron diffusion constant and $p_s$
is the momentum of a Cooper pair. The measured relaxation time, $F^*\tau_{Q^*}$,
is found to depend on $I_s$ through the relation, $F^*\tau_{Q^*}(I_s) = F^*\tau_{Q^*}(0)/(1+bI_s^2)^{1/2}$,
where $b$ is a function of $T/T_c$. The measured temperature dependence of $b$
is consistent with previous measurements of $1/F^*\tau_{Q^*}$ in AlEr films, in which
the Er is a pair-breaking magnetic impurity, and with the Schmid-Schön theory.
The measured magnitude of $b$ is in good agreement with the value expected
from measured sample parameters. As with the data on AlEr, the measured
value of $b$ is not consistent with results based on a numerical solution
of the Boltzmann equation.
I. INTRODUCTION

In an earlier paper\(^1\)(LC) we reported measurements of the charge-imbalance relaxation\(^2,3\) rate, \(1/F^*\tau_{Q^*}\), in Al doped with varying concentrations of a pair-breaking magnetic impurity, Er. The presence of the Er greatly enhances the relaxation rate because it allows branch crossing through elastic spin-flip scattering of quasiparticles.

LC compared their results with the Schmid-Schön\(^4\)(SS) prediction, which is valid near the transition temperature \(T_c\):

\[
\frac{1}{F^*\tau_{Q^*}} = \frac{\pi}{4\tau_E} \frac{\Delta}{k_B T_c} \left(1 + 2\frac{\tau_E}{\tau_S}\right)^{\frac{3}{2}} \quad (\tau_{Q^*}^{-1} \ll \tau_E^{-1}, \hbar^2\Gamma/\tau_E\Delta^2 \ll 1) \quad (1.1)
\]

Here \(\tau_E^{-1}\) is the electron-phonon scattering rate\(^5\) of an electron at the Fermi energy at \(T = T_c\), \(\tau_S^{-1}\) is the electron spin-flip scattering rate in the normal state\(^6\), \(\Gamma = (2\tau_E^{-1} + \tau_S^{-1})\), \(F^* \approx 1\), and \(\Delta(T)\) is the order parameter. In the temperature range \(0.2 \leq \Delta/k_B T_c \leq 1.4\) and for \(3 \leq \tau_E/\tau_S \leq 400\), the measured value of \(1/F^*\tau_{Q^*}\) was a universal function of \(\Delta/k_B T_c\). That is, the dependence on \(\Delta/k_B T_c\) was the same for all values of \(\tau_E/\tau_S\). Furthermore, \(1/F^*\tau_{Q^*}\) was proportional to \(\Delta/k_B T_c\) for \(\Delta/k_B T_c \leq 0.8\), and, at fixed temperature, was proportional to \((1 + 2\tau_E/\tau_S)^{\frac{3}{2}}\). Thus, although Eq. (1.1) is strictly valid only for \(\Delta/k_B T_c \ll 1\), it accurately describes the dependence of the data on \(\Delta/k_B T_c\) for \(\Delta/k_B T_c \leq 0.8\), and on \(\tau_E/\tau_S\) for all temperatures and values of \(\tau_E/\tau_S\) studied.
To compare rigorously their data with the theory at temperatures lower than the restricted range near $T_c$ in which the SS result is valid, LC solved the Boltzmann equation numerically for $1/F^*\tau_Q^*$. This solution agreed with the SS result, Eq. (1.1), for $\Delta/k_BT_c \ll 1$, but at higher values failed to reproduce the observed temperature dependence and universal behavior. This discrepancy between the observed and computed values of $1/F^*\tau_Q^*$ raised the question of whether the Boltzmann equation for charge relaxation failed for pair-breaking mechanisms in general or for magnetic impurities in particular. The chief objective of the present paper is to answer this question by studying charge relaxation in the presence of another pair-breaking mechanism; namely a supercurrent. The supercurrent induces charge relaxation because it raises quasiparticle energies by $\overrightarrow{p_k} \cdot \overrightarrow{v_s}$, where $\overrightarrow{p_k}$ is the momentum of the quasiparticle in state $k$, and $\overrightarrow{v_s}$ is the superfluid velocity. The induced asymmetry in the gap implies that the coherence factor for elastic non-spin-flip scattering from one branch to the other is no longer zero, as it is for a superconductor with an isotropic gap in the absence of a pair-breaking mechanism. Maki $^8$ has shown that the appropriate pair-breaking rate for thin films [d $\ll (\xi_0)^{-1}$] in the dirty limit ($\xi \ll \xi_0$) is

$$\frac{1}{\tau_s} = \frac{D}{2} \left( \frac{p_s}{\ell k_F} \right)^2,$$

where $\ell$ is the electron mean free path, $d$ is the film thickness, $\xi_0 = \hbar v_F / \pi \Delta(0)$, $D = v_F \ell / 3$ is the electron diffusion coefficient, $v_F$ is the Fermi velocity, and $p_s$ is the momentum of a Cooper pair.

The paper is organized as follows. In Sec. II we outline the theory of charge-imbalance generation and detection by tunneling, and describe how the SS theory and the Boltzmann equation used by LC can be adapted to describe charge relaxation in the presence of a supercurrent. Section III describes the experimental procedures and compares the experimental results with the
SS result and the numerical calculations. Section IV contains a summary and our conclusions.

II. THEORETICAL BACKGROUND

A detailed discussion of charge-imbalance generation and detection and of the Boltzmann equation used to compute the charge-relaxation rate can be found in Refs. 9 and 1. Here we give only the essential results.

We consider an experimental arrangement consisting of three metal films that form two overlapping tunnel junctions. When a current $I_i$ is passed through one junction, the injector, a steady-state charge imbalance is created in the middle film. The charge imbalance generates a voltage, $V_d$, that is detected by the second junction, the detector. The charge-relaxation rate is found from

$$
\frac{1}{F\tau Q^*} = \frac{I_i}{2N(0)e^2\Omega V_d g_{NS}(0)}
$$

(2.1)

where $N(0)$ is the single-spin density of states at the Fermi energy, $\Omega$ is the injected volume, and $g_{NS}(0)$ is the low-voltage conductance of the detector junction normalized to its value at $T_c$. Although, in general, both $F^*$ and $\tau Q^*$ depend on $V_i$, for the range of injection voltages used in the present experiment to determine $1/F^*\tau Q^*$, $10\Delta(T) \leq |eV_i| \leq 40\Delta(T)$, $1/F^*\tau Q^*$ was found to be independent of $V_i$.

Since LC found that for a given temperature $1/F^*\tau Q^*$ was proportional $(1 + 2\tau E/\tau_s)^{1/2}$, in agreement with the SS expression, we can combine this result with Eqs. (2.1) and (1.2) to obtain the expected dependence of $V_d$ on $I_s$:

$$
V_d(I_s) = \frac{V_d(0)}{(1+2\tau E/\tau_s)^{1/2}} = \frac{V_d(0)}{(1+b^{SS}I_s^2)^{1/2}}
$$

(2.2)
where, from Eq. (1.2),

\[
b^{ss} = \frac{2\tau E}{\tau_s I_s^2} = \frac{\tau E p_s^2}{\hbar^2 I_s^2} .
\]  

(2.3)

In these equations, \(V_d\) is the detected voltage for fixed \(I_i\) and \(\Delta/k_BT_c\), and \(I_s\) is the supercurrent. In the limit \(I_s^2 \ll I_c^2\), where \(I_c\) is the critical current, \(p_s\) is proportional to \(I_s\), so that \(b^{ss}\) is independent of \(I_s\). Using the familiar relations\(^{10}\)

\[
p_s = 2m\nu_s, \quad j_s = n_s e\nu_s, \quad n_s = mc^2/4\pi e^2\lambda^2, \quad \text{and} \quad \lambda(0) = \lambda_L(0)(1 + \xi_0/\xi) \approx \lambda_L(0)\sqrt{(\xi_0/\xi)} \quad \text{in the dirty limit, we find}
\]

\[
b^{ss} = \left(\frac{8\pi e^4}{\hbar c^2}\right)^2 \tau_E v_F^2 \lambda_L^4(0) \frac{\lambda^4(T/T_c)}{\lambda^4(0)} .
\]  

(2.4)

In Eq. (2.4), \(\lambda(T/T_c)/\lambda(0)\) is the penetration depth normalized to its value at \(T = 0\), \(\lambda_L(0)\) is the London penetration depth at \(T = 0\), \(m\) is the electronic mass, \(n\) is the density of conduction electrons, and \(w\) and \(d\) are the width and thickness of the film. In setting \(I_s = j_s wd\), where \(j_s\) is the supercurrent density, we have assumed a uniform current distribution. Note that the temperature dependence of \(b^{ss}\) is completely determined by \(\lambda^4(T/T_c)/\lambda^4(0)\), which in the dirty limit is a universal function of \(T/T_c\) given by Eq. (3.17) in Ref. 10.

We now turn to a brief discussion of the Boltzmann equation for a steady-state spatially uniform charge imbalance\(^{1,9}\):

\[
\frac{\partial f_{\varepsilon}}{\partial t} = G_{\varepsilon} - G_{\text{inc}} - G_{\text{ele}} - G_{\text{sfe}} - G_{\text{sce}} = 0 .
\]  

(2.5)

In this equation, \(f_{\varepsilon}\) is the quasiparticle distribution function, \(\varepsilon = \pm(E^2 - \Delta^2)^{1/2}\), where \(\pm\) refers to states above and below the Fermi energy, and \(E > 0\) is the excitation energy. The quantity \(G_{\varepsilon}\) is the net rate at which quasiparticles are injected into each state \(\varepsilon\); \(G_{\text{inc}}\) is the net rate at which quasiparticles scatter out of each state \(\varepsilon\) due to inelastic electron-phonon scattering; \(G_{\text{ele}}\), \(G_{\text{sfe}}\), and \(G_{\text{sce}}\) are...
is the net rate at which quasiparticles scatter from each state $\epsilon$ to all states $-\epsilon$ due to elastic non-spin-flip scattering in the presence of an intrinsically anisotropic energy gap; $G_{s\epsilon}$ is the net rate at which quasiparticles undergo elastic spin-flip scattering from each state $\epsilon$ to all states $-\epsilon$; and $G_{s\epsilon c}$ is the net rate at which quasiparticles scatter from each state $\epsilon$ to all states $-\epsilon$ due to elastic non-spin-flip scattering in the presence of gap anisotropy induced by the supercurrent. Expressions for $G_{\epsilon}$, $G_{inc\epsilon}$, and $G_{ele\epsilon}$ can be found in Ref. 9, and for $G_{s\epsilon}$ in Ref. 1. We neglect $G_{ele\epsilon}$ and $G_{s\epsilon}$ because in the present experiment the very short mean free path virtually eliminates the gap anisotropy and because we have not added magnetic impurities.

Using Green's functions, Bulyzhenkov and Ivlev\textsuperscript{11} obtained the result

$$G_{s\epsilon c} = \frac{D}{2} \left( \frac{p_s}{\hbar} \right)^2 \frac{2}{E_{\epsilon c}} \left( f_\epsilon - f_{-\epsilon} \right).$$

(2.6)

This term is identical to $G_{s\epsilon c}$ [LC Eq.(5.3)] if we use Eq. (1.2), so that all of the LC numerical results for charge relaxation in the presence of magnetic impurities are directly applicable to the present work. For fixed $\Delta/k_B T_c$, and for the range $0 \leq \tau_E/\tau_s \leq 1.6$ appropriate to this work, the LC numerical results predict that

$$V_d(I_s) = \frac{V_d(0)}{(1 + b_{\text{num}}/I_s)^{1/2}}.$$  

(2.7)

Equation (2.7) has the same form as Eq. (2.2), but $b_{\text{num}}$ differs from $b_{\text{ss}}$ as shown in Fig. 1. In Sec. III, we compare measured values of $b(T/T_c)$, $b_{\text{meas}}$, with $b_{\text{ss}}(T/T_c)$ and $b_{\text{num}}(T/T_c)$. 
III. EXPERIMENTAL PROCEDURES AND RESULTS

A. Procedures

We used a similar sample configuration to the one used by LC with the following changes (Inset, Fig. 2). The AlEr film was replaced with an Al film about 130 nm thick with the electron mean free path reduced by evaporation in an oxygen atmosphere. A supercurrent was applied to the Al strip via superconducting wires attached to the Pb tabs. To increase the uniformity of the supercurrent across the Al film, we sputtered a 500 nm Nb ground plane onto the substrate before depositing the sample. The Nb film was anodized and covered with about 200 nm of SiO and 30 nm of Ge. We believe that the current density was relatively uniform across the strip, except within ~300 nm of the edges, where it was somewhat higher. Furthermore, for the dirty films studied here, the penetration depth was comparable with the film thickness, so that the supercurrent should have been nearly uniform through the thickness of the film.

The electrical measurements were the same as those described by LC, except that at a number of temperatures in the range 0.7 to 0.9 \(T_c\), we measured \(V_d(I_s)\) as a function of supercurrent for fixed \(I_j\).

B. Results

We report results on the two best samples of the three in which we have observed an increase in \(1/F_*\) with increasing \(I_s\). The sample parameters are listed in Table I. The order parameter, \(\Delta\), followed the BCS \(^1\) temperature dependence to an accuracy of \(\pm3\%\) to within a few mK of \(T_c\). The normalized low-voltage conductance of the detector junction was larger than predicted \(^2\), but never by more than 10\%. This discrepancy does not lead to an error provided the experimental value is used in Eq. (2.1) to determine \(1/F_*\) \(^3\).
Measured values of $1/F^*\tau_Q^*$ vs. $\Delta/k_BT_c$ are shown in Fig. 2, along with theoretical curves obtained from Chi and Clarke\(^9\) (CC) and fitted to the data by using the values of $\tau_E$ and $\tau_{Qelo}^{-1}$ listed in Table I. We used the value\(^20\) $N(0) = 1.74 \times 10^{28}$ eV$^{-1}$m$^{-3}$ to obtain $1/F^*\tau_Q^*$ from Eq. (2.1). The quantity $\tau_{Qelo}^{-1}$ is the characteristic rate introduced by CC for branch relaxation by elastic non-spin-flip scattering, and it is proportional to the mean square gap anisotropy in pure Al, $\langle a^2 \rangle_0$. The fit is good for both samples, and yields values of $\tau_E$ consistent with those obtained by CC for films with similar transition temperatures. The values of $\tau_{Qelo}^{-1}$ yield values of $\langle a^2 \rangle_0$ within a factor of two of the expected value\(^21, 22\) of 0.01. Note that although branch crossing from elastic scattering does contribute to the shape of the plots in Fig. 2, the small value of $\tau_{Qelo}^{-1} = 0.03$ justifies our neglect of $G_{elc}$ in the derivation of $b_{num}$.

The largest values of $I_s$ attainable without driving the Al films normal were less than 2\%(6\%), for sample 1 (2), of the values of $I_c(T/T_c)$ estimated from the work of Skocpol\(^23\) on narrow Sn films. These estimates were made by using the value $I_c(0) = 23A [\text{Skocpol}^{23}, \text{Eq. (2)}]$, calculated from appropriate values of $H_c(0)$ and $\lambda(0)$, together with Fig. 1 of Ref. 23. The maximum attainable value of $\tau_E/\tau_s$ ranged from 0.1 (0.9) at $T/T_c = 0.78$ (0.70) to 0.01 (0.04) at $T/T_c = 0.91$ (0.89) for sample 1 (2). As $T$ increased above about 0.9 $T_c$, the maximum value of $I_s$ dropped abruptly to a value too small to have an appreciable effect on the detected voltage. We do not understand the reason for this sharp decrease.

A representative plot of $V_d(I_s)$ vs. $I_s$ for sample 2 is shown in Fig. 3. As predicted by Eq. (2.2) or (2.7), the value of $|V_d|$ decreased quadratically with increasing $|I_s|$ at low supercurrents, becoming nearly linear in $|I_s|$ at higher supercurrents. For sample 1, there was a slight asymmetry in the curves about $I_s = 0$ due to the non-negligible value of $I_1$ which also contributed to the charge relaxation. Thus, a function
with the form of Eq. (2.2) or (2.7) was fitted with the zero of \( I_s \) shifted by \( \pm I_i/2 \), depending on whether \( I_s \) and \( I_i \) added or subtracted in the junction area. At each temperature, curves such as those shown in Fig. 3 were fitted by eye for each combination of \( \pm I_i \) and \( \pm I_s \), and the four values of \( b \) were then averaged. The quality of the fits was excellent.

Experimentally determined average values of \( b \) are shown in Fig. 4, with error bars representing the spread in the four values of \( b \) obtained at each temperature and each value of \( I_i \). We looked for a dependence of \( b \) on \( V_i \) in sample 1 for \( 5\Delta \leq |eV_i| \leq 40\Delta \) and found none. The solid curves were obtained by scaling \( b^{ss} \) [Eq. (2.4)], to fit the data. The predicted temperature dependence is in excellent agreement with the data. The dashed curves are obtained by multiplying the solid curves by the ratio \( b^{num}/b^{ss} \) shown in Fig. 1, and rescaling to fit at \( T/T_c \approx 0.9 \). We see immediately that \( b^{num} \) is in substantial disagreement with the measured temperature dependence. In Table I we list values of \( <b^{ss}/b^{meas}> \) averaged over temperature, where \( b^{ss} \) is calculated using \( \lambda_L(0) = 16nm, v_F = 1.36 \times 10^6 m/s, \xi_0 = 0.18 \hbar v_F/k_B T_c, \) with the measured values of \( T_c, \tau_E, \xi, d, \) and \( w \). The ratio \( <b^{ss}/b^{meas}> \) varies between about 0.35 and 0.5. Given the uncertainties in the values of \( v_F, \xi_0, \lambda_L(0), \rho \xi, \) and \( \tau_E \), we consider the agreement to be very acceptable.

IV. SUMMARY AND CONCLUSIONS

We have measured \( F^{*} \tau_Q^{*} \) in dirty Al films in the presence of pair-breaking induced by an applied supercurrent, \( I_s \). The measured values of \( \tau_E \) for our films were consistent with those obtained by Chi and Clarke, and the values of \( \langle a_0^2 \rangle \) were within a factor of two of the expected value. We measured values of \( F^{*} \tau_Q^{*} \) as a function of \( I_s \) at fixed temperature and injection current in the range \( 0.7 \leq T/T_c \leq 0.9 \), and found that
$F^* \tau_Q^*$ was proportional to $1/(1 + b_{s}^2)^{1/2}$. The temperature dependence of $b_{\text{meas}}$ agreed well with that of $b^{ss}$ but not $b^{\text{num}}$, where $b^{ss}$ resulted from previous measurements on AlEr and $b^{\text{num}}$ resulted from a numerical solution of the Boltzmann equation. The average value, $<b^{ss}(T/T_C)/b^{\text{meas}}(T/T_C)>$, was $0.43 \pm 0.8$. We consider this to be satisfactory agreement in view of the uncertainties in the values of $v_F$, $\lambda_L(0)$, and $\rho_c$ taken from the literature, and in the measured value of $\tau_E$, all of which are required to calculate $b^{ss}$.

The temperature dependence of $b^{ss}$, and hence $b^{\text{meas}}$, was entirely consistent with our measurements of $1/F^* \tau_Q^*$ in AlEr, in which the pair breaking was due to magnetic impurities. As with the measurements on AlEr, the temperature dependence predicted by the numerical solution of the Boltzmann equation disagreed significantly with the data. We conclude that this disagreement involves the present understanding of the effect of pair breaking on charge-imbalance relaxation in general, and not the effect of any pair-breaking mechanism in particular. It is to be hoped that the source of this discrepancy will be investigated theoretically.

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References

5. Strictly, $\tau_\text{E}$ is a temperature dependent scattering time, defined in Ref. (4), Eq. (43). Since it is nearly constant for energies less than $\sim k_B T_c$, we use the symbol $\tau_\text{E}$ to represent this constant value.
14. This value of $v_F$ is the free electron value divided by the effective mass ratio, $m^*/m$, determined from the electronic specific heat coefficient, (See Ref. 16). It is consistent with the value for Al, $\xi_0 = 1.6 \mu m$, given in Ref. 17.


Table I. Measured and calculated parameters for samples 1 and 2.

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Film Thickness (nm)</th>
<th>Tc (K)</th>
<th>R300/R4.2</th>
<th>ρ4.2 (nΩm)</th>
<th>ℓ (nm)</th>
<th>Rj (Ω)</th>
<th>Rd (mΩ)</th>
<th>τE (ns)</th>
<th>τQeoνE</th>
<th>( &lt;a^2 &gt;_0 )</th>
<th>( \frac{b^{SS}}{b^{meas}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>113</td>
<td>1.370</td>
<td>1.89</td>
<td>35.6</td>
<td>25</td>
<td>0.3</td>
<td>3.4</td>
<td>5.3</td>
<td>0.031</td>
<td>0.005</td>
<td>0.36</td>
</tr>
<tr>
<td>2</td>
<td>147</td>
<td>1.514</td>
<td>1.43</td>
<td>81.3</td>
<td>11</td>
<td>1.3</td>
<td>6.9</td>
<td>2.7</td>
<td>0.031</td>
<td>0.018</td>
<td>0.51</td>
</tr>
</tbody>
</table>

R$_{300}$/R$_{4.2}$ is the residual resistivity ratio, ρ$_{4.2}$ is the resistivity of the Al film at 4.2 K, R$_j$ and R$_d$ are the resistances of the injector and detector junctions with all films normal, and ℓ is obtained from $ρ_{4.2}ℓ = 9 \times 10^{-16} \Omega \text{ m}^2$ (Ref. 13). We used $v_F = 1.36 \times 10^6 \text{ ms}^{-1}$ (Ref. 14) to deduce $<a^2>_0$, and took $\lambda_L(0) = 16 \text{ nm}$ (Ref. 16).
Figure Captions

Fig. 1. Plot of $b^{\text{num}}/b^{\text{ss}}$ vs. $T/T_c$.

Fig. 2. Plot of $1/F^* \tau_Q^*$ vs. $\Delta/k_B T_c$ for both samples. The solid lines are numerically calculated curves, fitted to the data by adjusting the parameters $\tau_E$ and $\tau_{Q\text{elo}}^{-1} \tau_E$. Inset shows the sample configuration.

Fig. 3. Typical experimental plot of $V_d$ vs. $I_s$ for fixed $I_i$. The points represent a fit "by eye" to a function of the form $V_d(I_s) = V_d(0)/(1 + b I_s^2)^{1/2}$, with $b$ as the fitting parameter.

Fig. 4. The data points are measured values of $b$, and the solid line is a fit of $b^{\text{ss}}$ to the data. The dashed curve is $b^{\text{num}}$ obtained from $b^{\text{ss}}$ using Fig. 1, and then scaled to fit the data at $T/T_c \approx 0.9$. Above $T/T_c \approx 0.9$, the dashed and solid curves are too close to draw separately.
Fig. 1

\[ \frac{b^{\text{num}}}{b^{ss}} \]

versus

\[ \frac{T}{T_c} \]

XBL808-5667
Sample No. 2

\[ \frac{T}{T_c} = 0.70 \]

\[ b = 3.35 \pm 0.1/b^2 \]

Fig. 3
Fig. 4

Sample No. 1

No. 2

XBL 808-5666

Fig. 4
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