Boundary resistance of the superconducting-normal interface

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The resistances of superconductor—normal-metal—superconductor sandwiches have been measured in which the mean free path of the superconductor was greater than or comparable with the coherence length. Below about 0.5 T_c, the resistance was nearly independent of temperature and, to within the experimental error, equal to the estimated resistance of the normal layer, indicating that there was relatively little interface contamination. At temperatures above about 0.9 T_c the resistance rose rapidly as the temperature was increased towards T_c. A theory for this rise, based on the normal metal-insulator-superconductor tunneling theory of Tinkham and Clarke, is used to calculate the quasiparticle charge imbalance, Q^*, injected into the superconductor from the normal layer. The resultant additional boundary voltage per unit current is expressed as a boundary resistance $R_b = Z(T)\,(D\tau_{Q^*})^{1/2}\rho_S/A$, where $Z(T)$ is a universal function of temperature, and D, τ_{Q^*}, ρ_S, and A are the electron diffusion coefficient, the charge relaxation time, the normal-state resistivity, and the cross-section area of the superconductor. Above 0.9 T_c, the data are an excellent fit to the theory if one takes $\tau_{Q^*} = 4k_BT\tau_{E=0}(T_c)/\pi\Delta_\infty(T)$, where $\tau_{E=0}(T_c)$ is the inelastic-scattering time at the Fermi surface at T_c, and $\Delta_\infty(T)$ is the energy gap far from the interface. The inferred values of $\tau_{E=0}(T_c)$ in $Pb_{0.99}Bi_{0.01}$, Sn, $Sn_{0.99}In_{0.01}$, and In, 0.25×10^{-10} s, 2.6×10^{-10} s, 1.1×10^{-10} s, and 1.1×10^{-10} s, respectively, are generally in good agreement with the computed values of Kaplan *et al.*

I. INTRODUCTION

Pippard, Shepherd, and Tindall[1] measured the resistance of superconductor—normal-metal—superconductor (S-N-S) sandwiches in which the normal metal was too thick and/or too dirty to sustain a Josephson supercurrent. They observed that, near the transition temperature of the superconductor, T_c, the resistance increased rapidly with increasing temperature. They ascribed this rise to the penetration of quasiparticles with energies greater than $\Delta_\infty(T)$ into the superconductor, where $\Delta_\infty(T)$ is the energy gap in S far from the interface. They also proposed that the additional boundary resistance was associated with a discontinuous jump in the electric potential at the N-S interface, the electric field being zero throughout the superconductor. However, Yu and Mercereau[2] showed that the potential did not fall abruptly to zero at the interface, but rather decayed exponentially in the superconductor. The work of Clarke and Tinkham[3–5] made clear that this potential in the superconductor arose from the presence of a quasiparticle charge imbalance.

$$Q^* = 2N(0)\int_\Delta^\infty (f_{k>} - f_{k<})\,dE_k \ , \qquad (1.1)$$

between the $k_>$ ($k > k_F$) and $k_<$ ($k < k_F$) quasiparticles. Here, $N(0)$ is the density of states per spin at the Fermi energy, f_k is the occupation number of the state k, and $E_k = (\Delta^2+\epsilon_k^2)^{1/2}$, where Δ is the energy gap and ϵ_k is the one-electron energy relative to the chemical potential. Subsequently, Harding, Pippard, and Tomlinson[6] studied the resistance of S-N-S sandwiches in which the mean free path of the superconductor was shortened by alloying, and found an additional boundary resistance at low temperatures as well as a greatly enhanced rise in resistance near T_c.

The N-S boundary resistance problem has been much studied theoretically. Rieger, Scalapino, and Mercereau[7] gave a description in terms of time-dependent Ginzburg-Landau[8] theory that contained some essentially correct ideas, but did not include considerations of charge imbalance or boundary scattering. The pioneering work of Pippard *et al.*[1]

and Harding *et al.*,[6] whose theory involved the solution of the Boltzmann equation, was extended by Waldram.[9] The microscopic theory was developed by Schmid and Schön,[10] and has been extended by Ovchinnikov,[11] Artemenko and co-workers,[12,13] and Krähenbühl and Watts-Tobin[14] (KWT).

The essential picture that emerges from this work is as follows. We consider first the case $l >> \xi_0$, where l is the electronic mean free path and ξ_0 is the BCS coherence length, and $\xi(T) << \lambda_{Q^*}$, where $\xi(T)$ is the Ginzburg-Landau coherence length and

$$\lambda_{Q^*} = (l v_F \tau_{Q^*}/3)^{1/2} \quad , \tag{1.2}$$

is the charge relaxation length in the superconductor.[4,5,10,15] In Eq. (1.2), v_F is the Fermi velocity in the superconductor, and τ_{Q^*} is the charge relaxation time[10,15] which, in general, contains contributions from both inelastic and elastic scattering processes (see Sec. II B). [Since the magnitude of the quasiparticle velocity is reduced by a factor $|\epsilon_k|/E_k$, strictly speaking one should use a suitable energy average of the velocity instead of v_F in Eq. (1.2). However, as we are concerned only with the limit $\Delta_\infty(T)/k_B T << 1$ in the theory developed in Sec. II, we shall neglect this correction.] Very close to $T_c (\Delta << k_B T)$ almost all of the excitations incident from N propagate into S, so that, in the presence of an external current, a quasiparticle current flows in the superconductor. In the usual situation where the transverse dimensions of the interface are much larger than the London penetration depth, there is no net current in the interior of the superconductor. The internal quasiparticle current is cancelled by a pair current, with a corresponding flow of supercurrent on the surface. The electric field is continuous at the interface, and the electric field, the electric potential, Q^*, and the quasiparticle current all decay exponentially into S with a characteristic length λ_{Q^*} (see Fig. 1). The boundary resistance is of order $\lambda_{Q^*}\rho_S/A$, where ρ_S is the normal-state resistivity of S, and A is the cross-sectional area of the interface.

When the temperature is lowered somewhat, a substantial fraction of quasiparticles have energies $\lesssim \Delta_\infty(T)$, and are Andreév reflected[16] at or near the N-S interface. In this process, a $k_>$ ($k_<$) quasiparticle incident from N is scattered onto the $k_<$ ($k_>$) branch, and the current carried by these two excitations continues in the superconductor as a supercurrent. Thus, there is no boundary resistance associated with these quasiparticles, and there is a discontinuity in the electric field at the interface (Fig. 1). The potential is continuous at the interface, but its spatial derivative is not. [In fact, the Andreév scattering process occurs over a distance $\sim \xi_0$, so that the discontinuities in the electric field and the derivative of the potential extend over this region. Furthermore (see below), there will be a small boundary resistance of order $\xi \rho_S/A \propto 1/l$ that is negligible in most practical situations.] In the presence of a current, the boundary scattering processes introduce disequilibrium in the quasiparticle distributions within an inelastic scattering length on either side of the interface.

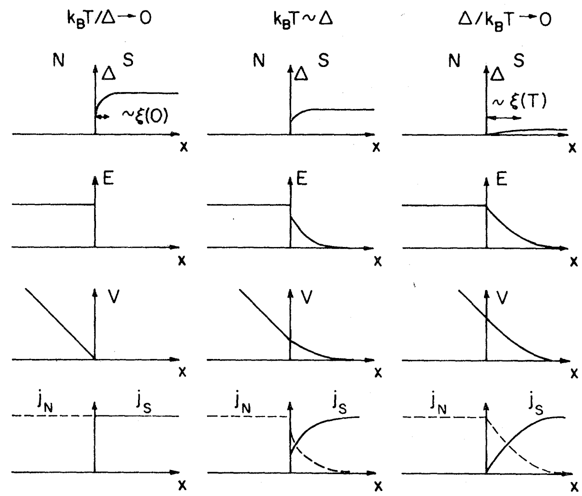

FIG. 1. Variation of energy gap, Δ, electric field, E, electric potential, V, normal current, j_N, and supercurrent, j_S, across an N-S interface for $k_B T/\Delta \to 0$, $k_B T \sim \Delta$, and $\Delta/k_B T \to 0$. The gap is taken to be zero in N.

As the temperature is lowered still further ($\Delta_\infty >> k_B T$), essentially all of the quasiparticles are Andreév[16] reflected at the interface, and there is no quasiparticle current in S. Correspondingly, the electric field and potential are zero in S (Fig. 1) and there is no boundary resistance (except for the small contribution mentioned above).

We now consider briefly the limit $l << \xi_0$. At low temperatures, the evanescent tail of the quasiparticles that extends into S for a distance $\sim (\xi_0 l)^{1/2}$ is subject to significant impurity scattering, as a result of which a substantial additional boundary resistance, of order $\rho_S(\xi_0 l)^{1/2}/A \propto 1/l^{1/2}$ is generated.[6,14] The existence of this contribution was convincingly demonstrated by Harding *et al.*,[6] and the magnitude and dependence on l were well explained by the theory of KWT.[14] Near T_c, Harding *et al.*[6] found a boundary resistance that increased with temperature, and, at a given temperature, with decreasing mean free path in the superconductor. KWT[14] explained this observation in terms of the *elastic* scattering of the quasiparticles in the region of length $\sim \xi(T) >> l$ in the superconductor over which $\Delta(T)$ varies with distance. In this situation,[4,5] elastic scattering may relax Q^*. KWT showed that, in a sufficiently dirty superconductor, the resistance due to this elastic scattering may completely dominate the resistance generated by inelastic relaxation of Q^*.

Although there appears to be reasonably good agreement between experiments in the extreme dirty limit and the KWT theory, there seems to have been no attempt to compare experiment and theory for relatively clean superconductors ($l \gtrsim \xi_0$), and to obtain estimates for τ_{Q^*} and hence $\tau_{E=0}(T_c)$, the inelastic scattering time at the Fermi energy at T_c. The present paper is concerned with this situation. Section II presents a simple theory that is appropriate in the limit $\Delta_\infty << k_B T$, and Sec. III describes the experimental procedures. The experimental results and their comparison with theory are discussed in Sec. IV, while Sec. V contains a summary.

II. THEORY

A. Tunneling model of N-S boundary resistance near T_c

In this section we develop a simple theory for the excess boundary resistance near T_c that fits our experimental data quite adequately, and enables us to deduce values of $\tau_{E=0}(T_c)$. We emphasize at the outset that this model is strictly valid only in the limit $\Delta_\infty << k_B T$, and that it is expected *a priori* to be seriously in error when $\Delta_\infty \sim k_B T$.

Figure 1 shows the variation of the superconducting order parameter, $\Delta(x,T)$, across an N-S interface near T_c. We assume that the transition temperature of the normal metal is much less than T, so that we can set $\Delta = 0$ for $x < 0$. In the superconductor, Δ rises from its value at the boundary, $\Delta_0(T)$, to its full value, $\Delta_\infty(T)$, over a distance of roughly the Ginzburg-Landau coherence length,[8] $\xi(T)$, that is always much less than λ_Q^* in the temperature range investigated experimentally. However, we note that quasiparticles with energies greater than $\Delta_\infty(T)$ may undergo some charge relaxation in the region where Δ varies spatially. At least in the limit $\Delta << k_B T$, this contribution to the overall relaxation rate is likely to be small, and we shall neglect it. We assume that the current densities are sufficiently low that they do not perturb Δ; the fact that the measured resistance is independent of current (see Sec. IV) suggests this is a good approximation. We further assume that quasiparticles with energies greater than $\Delta_\infty(T)$ are transmitted into S with probability unity; this is a reasonable approximation because of the relatively slow change of Δ with x. Quasiparticles with energies $< \Delta_\infty(T)$ are Andreév[16] reflected at a plane taken as $x = 0$ [since $\xi(T) << \lambda_{Q^*}$]. Finally, we assume that the quasiparticles are close to thermal equilibrium even in the vicinity of the interface; we emphasize that this is a reasonable approximation only for $\Delta_\infty << k_B T$.

Our calculation of the additional boundary potential (and hence the boundary resistance) is based on the model of Tinkham and Clarke[4] for tunnel injection into a superconductor, although we shall use the later Q^* formulation of Pethick and Smith.[15] Since the voltage across the interface is certainly small compared with $k_B T$, we can use the results appropriate for $eV << k_B T$. From Pethick and Smith[15] Eqs. (2.21) and (2.22), or, more immediately, from Clarke *et al.*[17] Eq. (6b), the charge imbalance generated by the uniform injection of a current I_{inj} into a volume Ω of a superconductor is given by

$$Q^* = \frac{Z(T)}{Y(T)} \frac{I_{\text{inj}}}{e\Omega} \tau_{Q^*} \quad (eV << k_B T) \ , \tag{2.1}$$

where

$$Z(T) = 2\int_\Delta^\infty N_S^{-1}(E) \left(-\frac{\partial f}{\partial E} \right) dE \ , \tag{2.2}$$

and

$$Y(T) = 2\int_\Delta^\infty N_S(E) \left(-\frac{\partial f}{\partial E} \right) dE \ . \tag{2.3}$$

Here, $N_S(E)$ is the normalized BCS density of states, f is the Fermi function, and $Y(T)$ is the BCS normalized conductance of a N-S tunnel junction in the limit $eV << k_B T$. In the present case, I_{inj} is just the quasiparticle current injected into S, and is related to the total current, I, by

$$Y(T) = I_{\text{inj}}/I \ . \tag{2.4}$$

Equation (2.4) follows from the realization that in a N-I-S tunnel junction at low voltages a fraction $1 - Y(T)$ of the current that flows at T_c cannot flow at a temperature $T < T_c$ because there are no states available in S at energies $< \Delta(T)$, whereas at the N-S interface, in our approximation, this same fraction $1 - Y(T)$ of the total current is transmitted into S as a pair current. Combining Eqs. (2.1) and (2.4), and replacing the exponentially decaying Q^* with a value that is constant at the value $Q^*(0)$ for $x \leq \lambda_{Q^*}$ and 0 for $x > \lambda_{Q^*}$, we find

$$Q^*(0) = \frac{Z(T) I \tau_{Q^*}}{eA\lambda_{Q^*}} \ . \tag{2.5}$$

The excess voltage, V_b, at each interface of the S-N-S sandwich adds to the voltage developed across the normal metal, and the total potential across the sandwich is measured with superconducting leads making metallic contact with the superconducting films. This is in contrast to the usual tunneling measurement[3] of Q^*, where the potential is measured by a tunneling contact to a normal metal. Thus, instead of the usual relation appropriate for the tunneling

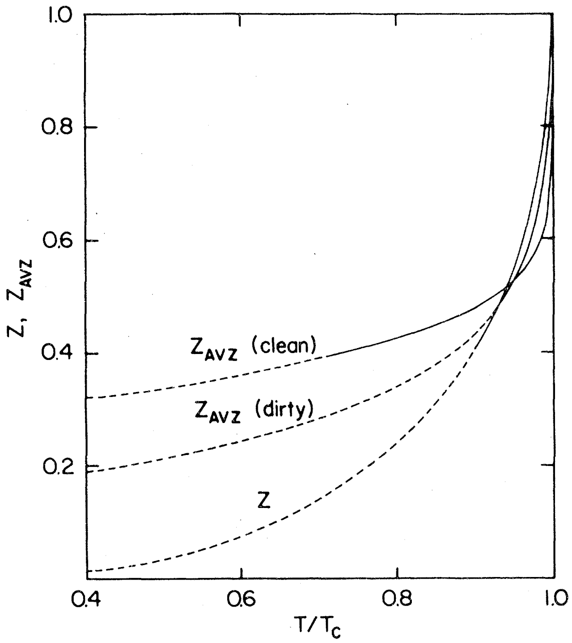

FIG. 2. $Z(T/T_c)$;$Z_{AVZ}(T/T_c)$ for Sn in the clean and dirty limits with $\tau_{E=0}(T_c)=2.7\times10^{-10}$ s, $\tau=5.5\times10^{-12}$ s. The solid lines indicate the range in which the theories are expected to be valid.

contact, $V=Q^*/2eN(0)g_{N-S}$, where g_{N-S} is the measured normalized conductance of the junction, we have

$$V_b=Q^*(0)/2eN(0) \quad . \tag{2.6}$$

Eliminating $Q^*(0)$ between Eqs. (2.5) and (2.6), and setting $\tau_{Q^*}=3\lambda_{Q^*}^2/lv_F$, we find a boundary resistance

$$R_b=\frac{V_b}{I}=\frac{Z(T)\lambda_{Q^*}\rho_S}{A} \quad , \tag{2.7}$$

where we have used the free-electron model[26] to calculate $\rho_S=3/2e^2N(0)lv_F$. $Z(T)$ is plotted versus T in Fig. 2.

Equation (2.7) is our final result, and it has a simple interpretation. As $T\rightarrow T_c$, $Z(T)\rightarrow1$, and the boundary resistance is just the resistance of a length λ_{Q^*} of the superconductor in the normal state. As the temperature is lowered, $Z(T)$ drops, reflecting the fact that fewer quasiparticles are able to propagate into the superconductor. At low temperatures[17]

$$Z(T)\approx(k_BT/\Delta)^{1/2}\exp(-\Delta/k_BT) \quad ,$$

so that R_b vanishes exponentially as $T\rightarrow0$, as we expect. However, despite the fact that Eq. (2.7) is a good approximation both near T_c and in the limit $T\rightarrow0$, we repeat our caution that it is *not* expected to be valid at intermediate temperatures where $\Delta_\infty\sim k_BT$.

B. Theory of Artemenko *et al.*

Artemenko, Volkov, and Zaitsev[13] (AVZ) have calculated the *S-N* boundary resistance, and find

$$R_b^{AVZ}=Z_{AVZ}(T)\rho_S\lambda_{Q^*}/A \quad , \tag{2.8}$$

where

$$Z_{AVZ}(T)\equiv\begin{cases}\left[1+0.68\dfrac{\Delta}{k_BT}\left(\dfrac{k_B\tau_{E=0}(T_c)T}{\hbar}\right)\right]^{-1} & \left[l\ll\xi_0,\ 1<\left(\dfrac{0.4\Delta^2\tau_{E=0}(T_c)}{\hbar k_BT_c}\right)^{1/4}<\dfrac{2k_BT}{\Delta}\right] , \quad (2.9a)\\[2ex] \left[1+0.66\left(\dfrac{\Delta}{k_BT}\right)^{1/2}\left(\dfrac{\tau_{E=0}(T_c)}{\tau}\right)^{1/8}\right]^{-1} & \left[l\gg\xi_0,\ 1<0.6\left(\dfrac{\tau_{E=0}(T_c)}{\tau}\right)^{1/8}<\dfrac{2k_BT}{\Delta}\right] . \quad (2.9b)\end{cases}$$

Here, τ^{-1} is the total electron scattering rate. Z_{AVZ} is plotted in Fig. 2 for the two limits using the value of $\tau_{E=0}(T_c)$ for Sn calculated by Kaplan *et al.*,[18] 2.7×10^{-10} s, and taking the value $\tau=5.5\times10^{-12}$ s appropriate for our Sn samples. As the temperature is lowered in the range near T_c, both forms of this theory drop off more rapidly than $Z(T)$.

C. Relation between τ_{Q^*} and $\tau_{E=0}(T_c)$

Assuming that one can extract a value of $\tau_{Q^*}(T)$ from the experimental data, one would like to derive from it a value for $\tau_{E=0}(T_c)$. Chi and Clarke[19] have computed $\tau_{Q^*}(T)$ versus temperature, and their low-voltage ($eV\ll k_BT$) result is plotted in Fig. 3 for Sn with $\tau_{E=0}(T_c)=2.7\times10^{-10}$ s.[18] In the range $T\gtrsim0.9T_c$ it is a reasonable approximation[19] (to within $\pm15\%$) to take

$$\tau_{Q^*}(T)=\frac{4}{\pi}\tau_{E=0}(T_c)\frac{k_BT}{\Delta}\ (T\lesssim0.9T_c,eV\ll k_BT) \quad . \tag{2.10}$$

Below $T/T_c\approx0.8$, $\tau_{Q^*}(T)$ increases rapidly as the temperature is lowered.

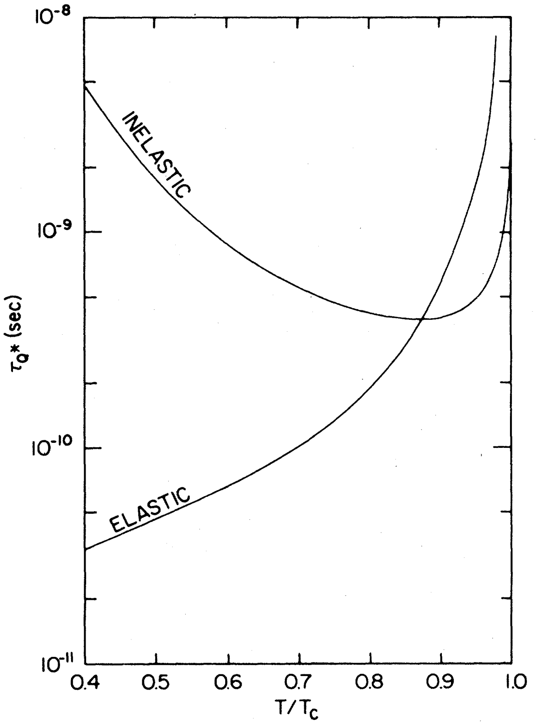

FIG. 3. Inelastic (taken from Ref. 19) and elastic [computed from Eq. (2.11)] charge relaxation times for $Sn_{0.99}In_{0.01}$. The elastic time is based on a simple model assuming all quasiparticles are at a temperature T, and is not meant to represent a detailed numerical calculation.

In the presence of an anisotropic energy gap, it is important to consider the contribution of elastic scattering to charge relaxation. Following Tinkham's model,[5] we can make the following rough estimate of $\tau_{Q^*\mathrm{el}}$ for quasiparticles at a characteristic temperature T[20]:

$$\tau_{Q^*\mathrm{el}} \approx \frac{\tau_1}{\langle a^2 \rangle_0}\left[1+\left(\frac{\hbar}{2\tau_1\Delta}\right)^2\right]\frac{k_B T}{\bar{\Delta}}\left(2+\frac{k_B T}{\bar{\Delta}}\right) . \quad (2.11)$$

In Eq. (2.11), τ_1 is the elastic scattering time, $\langle a^2 \rangle_0$ is the mean-square anisotropy for the clean superconductor,[23] and $\bar{\Delta}$ is the average gap. Equation (2.11) should be regarded as a rough estimate of $\tau_{Q^*\mathrm{el}}$ rather than a detailed calculation, but it should provide a sensible indication of the general behavior. In Fig. 3, we plot $\tau_{Q^*\mathrm{el}}$ for the $Sn_{0.99}In_{0.01}$ alloy used in our experiments (Table I), assuming[23] $\langle a^2 \rangle_0 = 0.02$, and $\tau_1 = 3.8 \times 10^{-13}$ s. It is evident that $\tau_{Q^*\mathrm{el}}^{-1}$ is negligible compared with the inelastic scattering rate for $T/T_c \gtrsim 0.9$, but increases rapidly as the temperature is lowered, becoming much greater than the inelastic rate at temperatures below about 0.8 T_c. A qualitatively similar behavior is expected for the other superconductors used in the experiments. In all cases, inelastic scattering dominates at temperatures above about 0.9 T_c while elastic scattering dominates at the lower temperatures.[29]

In principle, it should be possible to compute the combined inelastic and elastic charge relaxation rates at an arbitrary temperature, bearing in mind that these rates are strongly energy dependent and that the quasiparticle distribution is far from equilibrium for $\Delta_\infty \gtrsim k_B T$. In practice, this is likely to be a formidable task. It is a fortunate coincidence that, for temperatures above about 0.9 T_c, a simple model appears to be adequate to obtain τ_{Q^*} from the measured boundary resistance, and, in addition, τ_{Q^*} is dominated by inelastic scattering with the simple relationship to $\tau_{E=0}(T_c)$ given by Eq. (2.10). As $T \rightarrow 0$, $Z(T)$ become exponentially small so that Eq. (2.7) should again become valid, irrespective of the detailed behavior of τ_{Q^*}.

III. EXPERIMENTAL PROCEDURES

A. Sample preparation

We used two different types of S-N-S sandwiches. One was made by successive evaporations of the appropriate materials, while the other consisted of a normal-metal foil with a superconducting film evaporated onto each side. The properties of these samples are listed in Table I. To obtain samples with the desired properties it was necessary to use metals with low mutual solubilities that did not form intermetallic compounds at the temperature at which the samples were made. In addition, particularly in the case of the evaporated film samples, it was advantageous to shorten the electronic mean free path of the materials by adding impurities. This not only reduced the effects of interdiffusion and boundary disorder, but, also, in the case of the normal metal, helped to suppress Josephson supercurrents. Furthermore, λ_{Q^*} was reduced; as emphasized in Sec. II, it was necessary for this length to be much less than the thickness of the superconductor. However, we were careful not to make the superconductors so dirty that elastic scattering processes contributed significantly to the boundary resistance. With these considerations in mind, we made film samples from $Pb_{0.99}Bi_{0.01}$ and $Cu_{0.97}Al_{0.03}$ (Table I, column 1). Samples in which pure Sn was used and in which the thickness of the superconductor was comparable with λ_{Q^*} were unsatisfactory (Table I, column 5). In the case of the foil samples, the normal metal was Ir. It was impractical to alloy the Ir, but its mean free path appeared to be sufficiently shortened by intrinsic defects. We used Sn, $Sn_{0.99}In_{0.01}$, and In as the superconductors (Table I, columns 2, 3, and 4). Column 6 illustrates an attempt to make a sample using Pb on a Cu foil where

TABLE I. Properties of *S-N-S* sandwiches.

Sample	$Pb_{0.99}Bi_{0.01}$-$Cu_{0.97}Al_{0.03}$	Sn-Ir	$Sn_{0.99}In_{0.01}$-Ir	In-Ir	Sn-$Cu_{0.97}Al_{0.03}$	Pb-Cu
Type	Film	Foil	Foil	Foil	Film	Foil
Contact area, A (m^2)	(1) 3.5×10^{-8} (2) 3.7×10^{-8} (3) 1.9×10^{-8}	6.7×10^{-6}	6.7×10^{-6}	6.7×10^{-6}	2.4×10^{-8}	7.5×10^{-6}
T_c	7.16	3.73	3.74	3.41	3.73	7.19
Thickness of S (μm)	≈20	≈80	≈35	≈80	≈5	≈150
ρ_s ($\mu\Omega$ m)	2.18×10^{-2}	2.97×10^{-4}	4.23×10^{-3}	3.06×10^{-4}	3.20×10^{-4}	2.1×10^{-3}
mfp of S, l (μm)[a]	0.049	3.55	0.249	1.87	3.30	5.09
$\xi_0=\hbar v_F/\pi\Delta(0)$ (μm)[b]	0.076	0.24	0.24	0.44	0.24	0.075
l/ξ_0	0.64	14.8	1.04	4.25	13.8	67.9
Diffusivity of S, D (m^2s^{-1})[c]	7.8×10^{-3}	7.7×10^{-1}	5.4×10^{-2}	6.8×10^{-1}	7.2×10^{-1}	8.1×10^{-1}
Thickness of N, d_N (μm)	(1) 2.4 (2) 2.3 (3) 1.6	76	76	63	1.6	190
ρ_N ($\mu\Omega$ m)	1.91×10^{-2}	3.96×10^{-3}	3.96×10^{-3}	3.96×10^{-3}	1.91×10^{-2}	7.14×10^{-5}
$R_N=\rho_N d_N/A$ ($\mu\Omega$)	(1) 1.30 (2) 1.19 (3) 1.59	0.045	0.045	0.037	1.26	0.0018
R_0^{meas}	(1) 1.15 (2) 1.18 (3) 1.59	(1) 0.044 (2) 0.047 (3) 0.047	(1) 0.047 (2) 0.047 (3) 0.048	(1) 0.038 (2) 0.036	8.0	*d*
$\frac{1}{2}(R_0^{meas}-R_N)A$ ($10^{-15}\,\Omega\,m^2$)	(1) -2.6 (2) -0.2 (3) -0.1	(1) -3.4 (2) $+6.7$ (3) $+7.0$	(1) $+6.7$ (2) $+6.7$ (3) $+8.6$	(1) $+3.4$ (2) -3.4	80	*d*

[a]Estimated from $\rho_s l = 1.06\times10^{-15}$ $\Omega\,m^2$(Pb), 1.05×10^{-15} $\Omega\,m^2$(Sn), and 5.7×10^{-16} $\Omega\,m^2$(In). Values for Pb and Sn taken from Ref. 24 and for In from Ref. 25.

[b]Values of v_F were calculated from $v_F=\pi^2k_B^2/e^2\gamma\rho_s l$ (Ref. 26), where γ is the coefficient of electronic specific heat. Values of γ were taken from Ref. 27. $\Delta(0)=1.76\,k_BT_c$ (In,Sn), $2.15\,k_BT_c$ (Pb) (Ref. 28).

[c]Determined from $D=\pi^2k_B^2/3e^2\rho_s\gamma$ (Ref. 26).

[d]Because of interface contamination, no useful estimate of R_0^{meas} was possible.

the interface was apparently contaminated by an oxide layer. Thus, columns 1–4 represent "good" samples the data from which were analyzed in detail, whereas columns 5 and 6 represent "bad" samples, which illustrate various pitfalls in the sample preparation.

The Ir or Cu foils were prepared using a technique similar to that of Harding *et al.*[6] After chemical cleaning, the foils were clamped between two water-cooled aluminum masks containing concentric apertures, and sputter etched in argon at 50 kV at ~1 mA cm^{-2} for 30 min. The argon was pumped out of the vacuum system, and the Sn or In evaporated at ~100 nm s^{-1} onto each side of the foil in turn. Leads of the same material were spot welded to the disks of Sn or In. We made the film sandwiches by evaporating onto a water-cooled glass substrate a strip of superconductor, a disk of normal metal, and a second strip of superconductor perpendicular to the first. The initial pressure in the evaporator was typically 10^{-8} Torr, and the evaporation rate was about 100 nm s^{-1} for the superconductor and 10 nm s^{-1} for the Cu. Superconducting current and voltage leads were attached to the superconducting strips with Pb-Bi eutectic solder. For both types of sample, a separate strip of each evaporated material was deposited simultaneously, and used to determine the resistivity and transition temperature. Below T_c, the normal-state resistivity of all the superconductors, determined with the films driven normal by a magnetic field, was independent of temperature. The film thicknesses were measured with a quartz crystal monitor, and later checked with a Dektak[30] machine. Separate experiments were performed to ensure that the evaporated alloys were homogeneous.

B. Measurement technique

The samples, usually two or three at a time, were connected in series with a known resistor and the superconducting input coil of a dc SQUID (superconducting quantum-interference device) with a conventional feedback arrangement.[31] The samples were mounted on a copper plate in a vacuum can surrounded by a Pb shield and immersed in liquid helium. The temperature of the copper plate was regulated at the desired value. The resistance of each sample was measured as a function of current over the temperature range from about 1.2 K to T_c. The sensitivity of the measurements was limited by the Johnson noise in the samples and series resistor.

IV. EXPERIMENTAL RESULTS

We first describe the results obtained on the "good" samples (columns 1–4 in Table I), compare them with theory, and obtain values of $\tau_{E=0}(T_c)$. Second, we briefly discuss the behavior of the "bad" samples (columns 5 and 6).

A. Results obtained from "good" samples (Table I, columns 1–4)

The resistances of the samples were independent of current (to within ±1%) for current densities up to 3×10^6 A m^{-2}. The variation of resistance with temperature for one representative sample of each type is shown in Fig. 4. Near T_c, the resistances increase rapidly with increasing temperature, while at low temperatures the resistances are nearly independent of temperature. The measured asymptotic low-temperature values, R_0^{meas}, are shown in Table I, together with the values estimated from the measured resistivity, thickness, and area of the normal layer, R_N. The "excess" resistance of unit area for *each S-N* interface, $\frac{1}{2}(R_0^{\text{meas}}-R_N)A$, is also shown in Table I; it is negative for the PbBi samples, positive for the SnIn samples, and both positive and negative for the Sn and In samples. By way of comparison, Harding *et al.*[6] found a minimum value of $+2\times10^{-15}$ Ω m^2 for their samples. Given that R_0^{meas} sometimes exceeds and sometimes falls below R_N, we have little choice but to conclude that the low-temperature boundary resistance is zero, to within our experimental error. Since the thickness of the normal metal is typically hundreds of mean free paths, this result does not preclude the possibility of considerable disorder over several mean free paths, but it does indicate that there is not a significant oxide layer at the interface (see Sec. IV D). It is also noteworthy that for our dirtiest sample, PbBi, the predicted excess low-temperature boundary resistance due to the elastic scattering of sub-gap quasiparticles (see Sec. I) is of order $\rho_S(\xi_0 l)^{1/2}\sim10^{-15}$ Ω m^2, a value that is less than our experimental error in $\frac{1}{2}(R_0^{\text{meas}}-R_N)A$.

B. Comparison of data with theory

To compare our data with Eq. (2.7), in Fig. 5 we plot the total measured resistance versus $Z(T)\times(k_BT/\Delta)^{1/2}$ for each sample shown in Fig. 4. The solid lines represent a least-squares fit to the data for $T>0.9\,T_c$ (0.96 T_c for Sn). For PbBi, SnIn, and In the fit of the data above 0.9 T_c to a straight line is excellent, whereas for Sn the data show a slight curvature at temperatures below about 0.96 T_c. The reason for this different behavior is not clear. However, since the theory is strictly valid only for

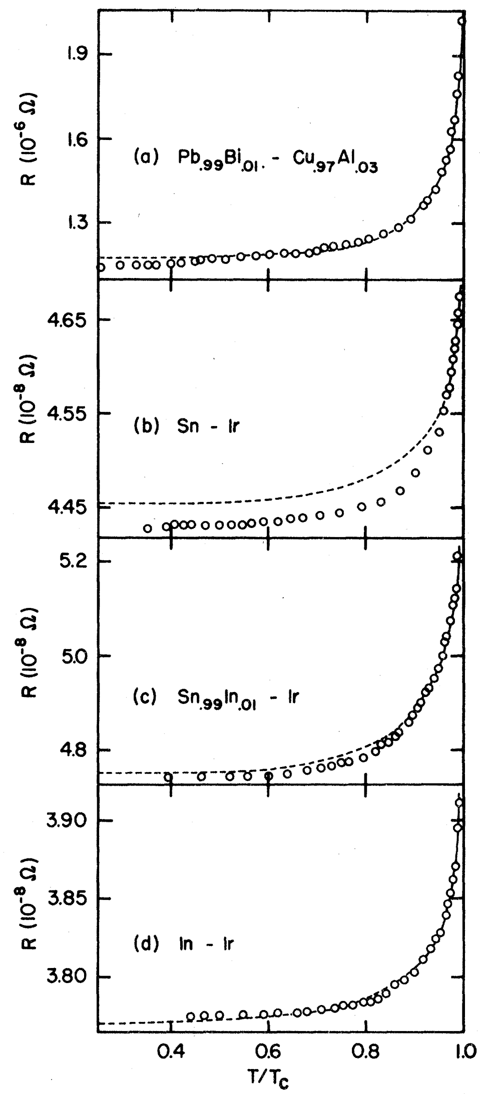

FIG. 4. Circles are measured dependence of the total resistance vs reduced temperature of one sample of each type listed in columns 1–4 of Table I. Samples shown are PbBi (2), Sn (1), SnIn (2), and In (1) (the numbers in parenthesis refer to the samples listed in Tables I and II). The solid lines above 0.9 T_c (0.96 T_c for Sn) are the fit to Eq. (2.7), and the dashed lines show the extrapolation of the theory to lower temperatures. The theory is *not* expected to be valid at intermediate temperatures.

TABLE II. Experimental values of $\tau_{E=0}(T_c)$, together with computed values from Kaplan *et al.* (Ref. 18). In the case of alloy samples, the computed values for the pure materials are given in parenthesis.

Material	$Pb_{0.99}Bi_{0.01}$	Sn	$Sn_{0.99}In_{0.01}$	In	Pb
$\tau_{E=0}(T_c)$ (10^{-10} s) (Experimental)	1. 0.24	2.6	1.18	1.06	0.22
	2. 0.22	3.2	1.05	1.06	0.19
	3. 0.30	2.0	1.12	...	...
	Average 0.25	2.6	1.1	1.1	0.20
$\tau_{E=0}(T_c)$ (10^{-10} s) (Kaplan *et al.*)	(0.23)	2.7	(2.7)	1.0	0.23

$\Delta \ll k_B T$, it is hardly surprising that small deviations from the experimental results occur for $\Delta \gtrsim 0.6\, k_B T$. Rather, it is somewhat surprising that the theory fits the data as well as it does for $\Delta \lesssim k_B T$ for the PbBi, SnIn, and In samples. From Fig. 5 we conclude that Eq. (2.7) is an adequate description of the variation of the resistance with temperature near T_c. The slope of the lines in Fig. 5 is $(4/\pi)^{1/2}\rho_S D^{1/2}\tau_{E=0}^{1/2}(T_c)/A$, and yields the values of $\tau_{E=0}(T_c)$ shown in Table II, where we have listed the values for all "good" samples, together with their average. The values obtained by Kaplan *et al.*[18] are also shown; we have converted their characteristic time, τ_0, to $\tau_{E=0}(T_c)$ using $\tau_{E=0}(T_c) = \tau_0/8.4$. (The values in parenthesis are those computed for the pure metals.) Our experimental values are generally in rather good agreement with the computed values.

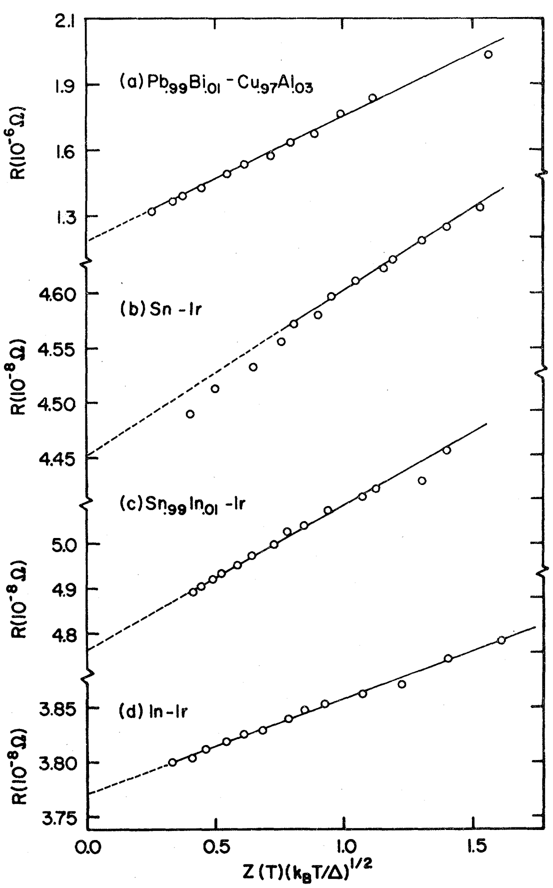

FIG. 5. Total measured resistance vs $Z(T)\,(k_B T/\Delta)^{1/2}$ for the samples shown in Fig. 4. The solid lines are a least-squares fit to the data for $T > 0.9\,T_c$ ($0.96\,T_c$ for Sn).

Using the appropriate values of $\tau_{E=0}(T_c)$ listed in Table II, we have plotted the resistance predicted by Eq. (2.7) in Fig. 4. As pointed out in Sec. II C, Eq. (2.7) is expected to be valid at low temperatures as well as near T_c. Except for Sn, the extrapolated low-temperature resistance is in good agreement with the measured resistance, thus providing a good check on the consistency of our results. Particularly in the cases of PbBi and In, the fit is remarkably good even at intermediate temperatures, a result that we believe to be coincidental, particularly since the expression used for τ_{Q^*}, Eq. (2.10), is quite inappropriate in this range. However, it may be that the increase in the elastic charge relaxation rate as the temperature is lowered tends to compensate for the decrease in the inelastic rate, thus keeping τ_{Q^*} roughly constant at temperatures below about $0.9\,T_c$.

C. Comparison of data with theory of Artemenko *et al.*

In Fig. 6 we have plotted the total sample resistance versus $Z_{AVZ}(k_B T/\Delta)^{1/2}$ in the range $T > 0.9\,T_c$ for the four samples shown in Fig. 4. We have used Eq. (2.9a) (dirty limit) for PbBi, and Eq. (2.9b) (clean limit) for the remaining materials. It is evident that the data lie on a curve throughout the temperature range. Unfortunately, our attempts to obtain an asymptotic slope in the limit $T \to T_c$ using a polynomial fit have not been successful because the data are too few and too scattered in this region. We have therefore been unable to extract values of $\tau_{E=0}(T_c)$ using this theory. It appears that Eq. (2.7) provides a rather better fit to the data than Eq. (2.9) for reasons that are not entirely clear. However, we note that as the temperature is lowered the experimental resistances in Fig. 6 drop off more rapidly than the corresponding values of $Z_{\rm AVZ}(T)/(k_BT/\Delta)^{1/2}$. Given the relatively good fit of the data to Eq. (2.7) shown in Fig. 5, this behavior is expected from Fig. 2, where one observes that $Z_{\rm AVZ}(T)$ (clean and dirty) drops off less rapidly than $Z(T)$ below about 0.95 T_c. It thus appears that, at least between 0.9 and 1.0 T_c, our neglect of the nonequilibrium corrections in the derivation of Eq. (2.7) is less serious than the fact that Eq. (2.9) is valid only to first order in Δ/k_BT. In fact, apart from

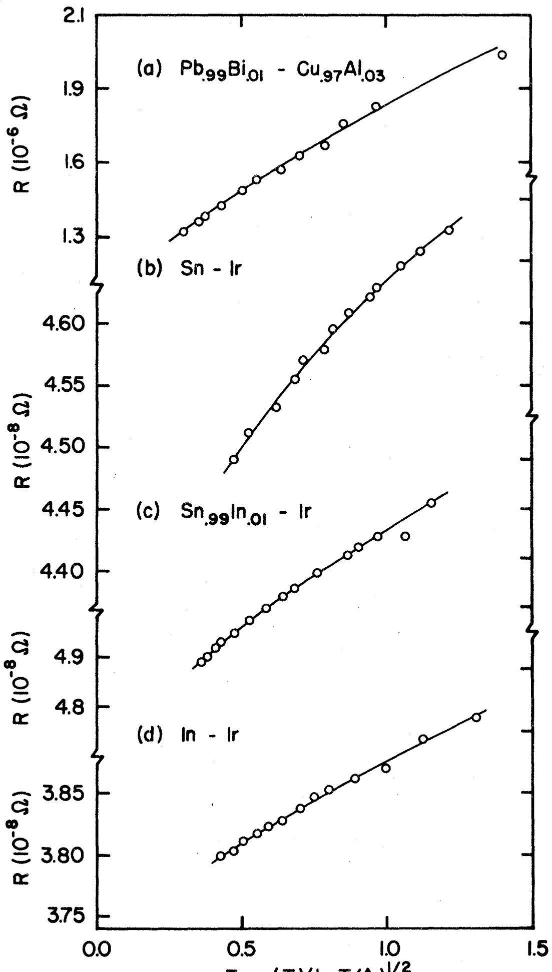

FIG. 6. Total measured resistance vs $Z_{\rm AVZ}(k_BT/\Delta)^{1/2}$ for the samples shown in Fig. 4. The curved lines are drawn through the data points, and indicate the relatively poor fit to the AVZ theory compared with the fit in Fig. 5.

a numerical factor, one could regard either of the $Z_{\rm AVZ}(T)$ as a first-order approximation to $Z(T)$, which, near T_c has the form[17]

$$1-\pi\Delta/4k_BT+[7\zeta(3)/4\pi^2](\Delta/k_BT)^2\cdots \quad .$$

It would be of interest to carry through the AVZ calculation to higher order, and to compare the results with the experimental data.

D. Results obtained from "bad" samples (Table I, columns 5 and 6)

We briefly discuss two samples illustrating difficulties with preparation that must be avoided to obtain "good" samples. Column 5 in Table I lists parameters for a $\rm Sn\text{-}Cu_{0.97}Al_{0.03}$ thin-film sample, and the temperature dependence of its resistance is shown in Fig. 7. Two problems are apparent. The measured resistance at low temperatures is much higher than R_N, corresponding to an excess resistance of 80×10^{-15} $\Omega\,\rm m^2$ at each boundary. This excess presumably arose from interdiffusion and alloying of the Sn and Cu.[32] At low temperatures, the dashed line in Fig. 7 represents R_N calculated from the measured parameters of the CuAl, while the rise at high temperatures is that expected from Eq. (2.7) using the measured value of ρ_S and $\tau_{E=0}(T_c)=2.7\times10^{-10}$ s. The mea-

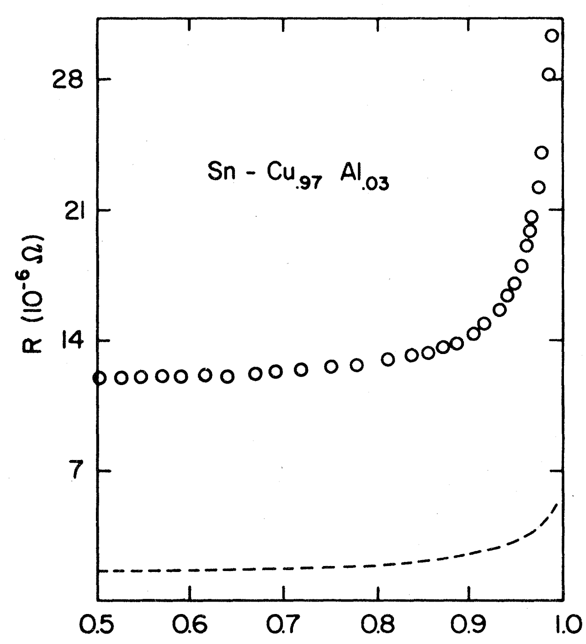

FIG. 7. Measured resistance (circles) vs T/T_c for Sn-CuAl (film) sample of Table I, column 5, in which substantial alloying has occurred and the Sn film is thinner than λ_{Q^*}. Dashed line shows predicted behavior in absence of alloying and for a thick Sn film.

sured rise near T_c is much greater than expected, perhaps partly because the Sn is contaminated by Cu, but largely because the thickness of the Sn, d_S, (5 μm) was much less than λ_{Q^*}, roughly 15 μm at 0.9 T_c. It is easy to show that in this limit the value of R_b is given by Eq. (2.7) should be enhanced by a factor λ_{Q^*}/d_S. It is impractical to obtain useful information from this sample.

The second "bad" sample is Pb-Cu (foil), shown in the last column of Table I and in Fig. 8. As the temperature was lowered from T_c, the resistance dropped, reached a minimum between 0.7 and 0.8 T_c, and then increased again. Harding *et al.*[6] observed similar effects, and suggested that a certain fraction of the *S-N* interface was contaminated with an insulating layer, for example an oxide, that formed a tunnel junction between the two materials. Near T_c, the normalized conductivity of the oxide patches is close to unity, and the contamination should have little effect on the resistance, but at lower temperatures the normalized conductivity of the patches drops, thereby increasing the measured resistance. Despite this problem, one should still be able to fit the data above 0.9 T_c, where the effects should be negligible. The values so obtained for $\tau_{E=0}(T_c)$ (Table II) are not significantly different from those obtained for PbBi.

Despite the fact that the values of $\tau_{E=0}(T_c)$ obtained from this sample were in good agreement with values obtained from samples with clean interfaces, a significant level of interface contamination is clearly undesirable. Since we were unable to obtain Cu foil samples without a resistance minimum, we used Ir instead for the normal layers. These samples showed no evidence of a resistance minimum, and had low-

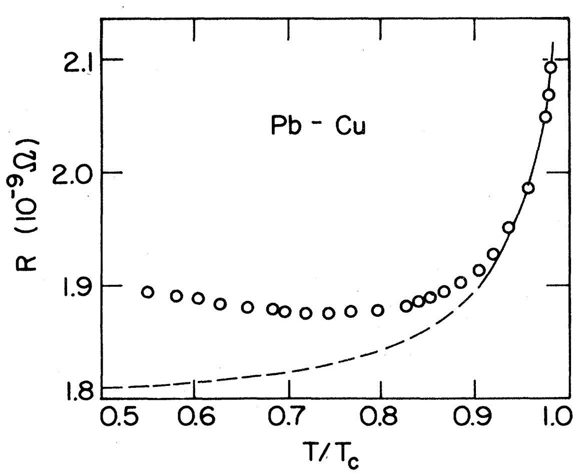

FIG. 8. Measured resistance vs T/T_c for Pb-Cu (foil) sample of Table I, column 6, showing effects of boundary contamination. Solid line is a fit to Eq. (2.7), and dashed line is the extrapolation of the theory to low temperatures.

temperature resistances close to those expected from the properties of the Ir, leading us to believe that the interfaces were relatively free from contamination.

V. CONCLUDING SUMMARY

We have fabricated *S-N-S* sandwiches in which the good agreement of the measured low-temperature resistance with the expected resistance of the normal metal and the lack of a resistance minimum suggests that the interfaces are relatively free from contamination. However, the resolution of these measurements is such that the existence of disorder over distances of several mean free paths from the interfaces cannot be ruled out. The rise in resistance as the temperature is increased towards T_c, ascribed to the propagation of a quasiparticle current into the superconductor, appears to be adequately fitted by a simple model adapted from the case of tunneling injection into a superconductor from a normal metal. This model is strictly valid only when $\Delta_\infty(T) << k_B T$, so that the departure of quasiparticles from equilibrium can be neglected, but in practice gives a good fit to the data down to 0.9 T_c where $\Delta_\infty(T) \approx k_B T$. Values of τ_{Q^*} inferred from this fit produced values of $\tau_{E=0}T_c)$ that are in good accord with the computed values of Kaplan *et al.*[18] The value of the resistance of the sandwich at $T=0$ extrapolated from the high-temperature fit is generally in good agreement with the measured low-temperature resistance. However, as emphasized earlier, the apparently reasonable agreement between experiment and theory at intermediate temperatures, where $\Delta_\infty(T) \lesssim k_B T$, is coincidental, since both the theory and the expression used for τ_{Q^*} are quite inapplicable in this range. Although one could, in principle, attempt to fit the data to more detailed theories, for example, that of Waldram,[9] at intermediate temperatures, the difficulty of accounting for both elastic and inelastic scattering processes and their strong energy dependence makes this a formidable undertaking.

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[20]Equation (2.11) is based on Eq. (50) of Ref. 6. However, as pointed out by J. Clarke and J. L. Paterson [Ref. 21, Eq. (16)], the Anderson averaging term, $1+(\hbar/2\tau_1\overline{\Delta})^2$ (Ref. 22), should not be squared. Furthermore, if we assume a typical electron energy in the superconductor is $\overline{\Delta}+k_BT$, and we make this substitution in Eq. (46) of Ref. 6, we arrive at Eq. (2.11).

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[29]It should be realized that the behavior indicated in Fig. 2 holds only for the case $eV \ll k_BT$. Thus, for the case of high-energy injection into Sn and Sn-In studied in Refs. 3 and 21 the measured value of τ_{Q^*} was more or less independent of temperature between about 0.8 T_c and 0.3 T_c. This is because the quasiparticles were injected at energies much greater than Δ, so that substantial inelastic charge relaxation occurred before the quasiparticles had thermalized.

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