Impurity scattering time in aluminum

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Using Gorkov's extension of the Ginzburg-Landau equations, we derive a formula for determining the impurity scattering time for a metal in the normal state in terms of the critical current density and transition temperature of the metal in the superconducting state. When we apply this formula to aluminum weak-link data, we find that the impurity scattering time varies by three orders of magnitude in going from clean to granular aluminum films. This corresponds quite well to impurity scattering times obtained directly from resistivity measurements of the films using the classical Drude-model formula.

I. INTRODUCTION-THEORY

The motion of an electron in a metal with randomly distributed impurities can be characterized by an impurity scattering time τ . In the Drude model the scattering time is related to the normal-state resistivity ρ_N through

$$\tau = m/Ne^2 \rho_N \quad , \tag{1}$$

where N is the volume density of electrons, m and e the mass and charge of the electron.

Using an extension of the Ginzburg-Landau (GL) theory, Gorkov² found the penetration depth λ_G in a superconductor is related to the impurity scattering time τ by

$$\lambda_G(T) = \lambda_L(T) / \sqrt{\chi(\rho)} \quad . \tag{2}$$

where, with the use of Gorkov's notation, $\rho = \hbar/2\pi\tau k_B T_c$, and $\chi(\rho)$ is given by

$$\chi(\rho) = \frac{\pi^2}{7\zeta(3)\rho} \left\{ 1 + \frac{4}{\pi^2 \rho} \left[\psi(\frac{1}{2}) - \psi(\frac{1}{2} + \rho) \right] \right\} . \quad (3)$$

Here, $\psi(\rho)$ is the logarithmic derivative of the Γ function, $\zeta(3) = 1.202$, T_c is the transition temperature of the superconductor and $\lambda_L(T)$ is the London penetration depth given by³

$$\lambda_L(T) = \left(\frac{mc^2}{4\pi Ne^2}\right)^{1/2} \frac{1}{[2(1-T/T_c)]^{1/2}} . \tag{4}$$

Therefore, from the penetration depth λ_G and with values of T_c and N known, τ can be directly calculated using Eqs. (2)-(4).

Near T_c and for microbridge widths and thickness smaller than the coherence length and the penetration depth, the penetration depth is related to the

critical current J_c of a thin-film microbridge by³

$$J_c(T) = \frac{cH_c(T)}{3\sqrt{6}\pi\lambda_G(T)} \bigg|_{T \to T_c} = J_c(0) \left[1 - \frac{T}{T_c}\right]^{3/2} , \quad (5)$$

where $H_c(T)$ is the critical magnetic field. With the use of the BCS results⁴

$$H_c(0) = \left[4\pi N(0) \frac{8(\pi k_B T_c)^2}{7\zeta(3)} \right]^{1/2}$$
 (6)

and

$$\lambda_L(0) = \left(\frac{mc^2}{4\pi Ne^2}\right)^{1/2} \tag{7}$$

and the large ρ approximation² for

$$\chi(\rho) = \frac{\pi^2}{\rho \to \infty} \frac{\pi^2}{7\zeta(3)\rho}$$

in Eq. (5) (in the free-electron approximation)

$$\tau = \frac{2}{\pi} \left[\frac{63\zeta(3)}{32} \right]^2 \frac{\hbar^7}{e^2 m^2 k_B^3} \frac{J_c^2(0)}{E_f^2 T_c^3} , \qquad (8)$$

where N(0) is the density of states per unit volume of electrons at the Fermi surface of one spin and E_F is the Fermi energy. Therefore the scattering time τ can be determined from measurements of J_c and T_c , along with free-electron values for E_F .

We have determined the scattering times for aluminum as described above for a range of samples for which τ varied by three orders of magnitude. The scattering times obtained from independent resistivity measurements in the normal state are in reasonable agreement with the ones calculated from measurements of the critical current of microbridges.

II. EXPERIMENTAL DETAILS:

Aluminum films 300-6000 Å thick were deposited onto sapphire substrates using an electron beam gun at rates of 5-80 Å/sec in background pressures of 2×10^{-6} to 2×10^{-5} Torr. High evaporation rates at low pressures were used to obtain clean films while low rates at high pressures produced dirty films. Using standard photolithographic techniques, a series of six microbridges of fixed 10-µm length but varying widths ranging from 0.50 to 2.5 μ m were etched on each substrate. By varying the deposition rate and the partial pressure of oxygen, we were able to produce films with T_c 's ranging from 1.24 to 2.35 K and resistivity ratios ranging from 12 to 1, respectively. Film thicknesses were measured both by a quartz crystal monitor and with a Dektak.⁵ After the measurements, the width of each bridge was determined to within 0.1 µm using a scanning electron microscope.

The current and voltage leads were attached to the samples using Be-Cu pressure contacts. The temperature of the helium bath was controlled using a feedback configuration of a calibrated Ge thermometer and a heater, while the absolute temperature was measured with a separate calibrated Ge thermometer. The measurements were made in a Dewar surrounded with μ -metal shielding and in a filtered-screen room environment. By filtering all four current and voltage leads with low-pass L-C filters and the voltage leads with high-pass R-C filters, the system had a noise temperature equal to the bath temperature.

The differential resistance (dV/dI) versus current

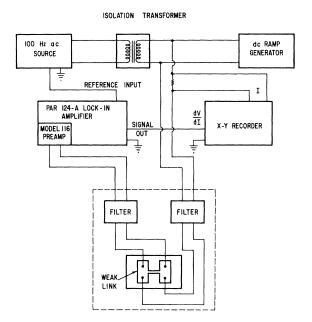


FIG. 1. Low-noise, phase-sensitive technique for measuring dV/dI vs I in weak-link microbridges.

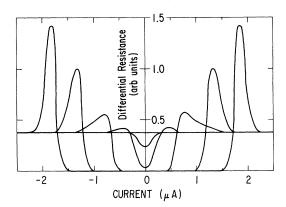


FIG. 2. dV/dI vs I curves spaced at 1-mK temperature increments for a $10 \times 1.5 \times 0.1$ - μ m³ bridge. The horizontal line curve gives a T_c of 1.230 K.

(I) of the microbridges at each temperature was measured using a phase sensitive technique as shown in Fig. 1. A 100-Hz, $0.1-\mu A$ rms signal was applied across the microbridge and at the same time a dc bias current was ramped up. The resulting modulated voltage was detected with a lock-in amplifier. Figure 2 depicts a series of dV/dI vs I curves of an aluminum microbridge for various temperatures near T_c . When the ratio of the energy associated with the critical current $\hbar I_c/e$ is larger than the thermal energy $k_B T_c$, i.e., $\gamma = \hbar I_c / e k_B T_c > 10$, then I_c can be directly determined from the position of the peak in the dV/dI vs I curve. When $\gamma \leq 10$, there exists a finite resistance across the microbridge even at zero-bias current, and the theory of Ambegaokar-Halperin⁷ is needed to calculate I_c . The transition temperature T_c is the temperature at which all structure in the differential resistance vanishes.

III. EXPERIMENTAL RESULTS

Figure 3 shows the mean-field plot $I_c^{2/3}$ vs T for a series of microbridges of equal thickness but different widths. The temperature dependence of I_c is in good agreement with Eq. (5) except in the vicinity of T_c for dirty films where there is a "tail" in the data. The origin of this tail has been the subject of an earlier paper. 8 T_{c} is determined experimentally as the point where the critical current is identically zero. A universal curve (Fig. 4) for all microbridges can be obtained by normalizing critical currents to the extrapolated zero-temperature value $I_c(0)$ and using the straight line extrapolation to $I_c = 0$ to find T'_c . The data follow the GL result up to a $T/T_c' = 0.99$, at which point the data roll off toward values of T/T_c greater than 1. In all cases the value of T_c' determined from extrapolation of the mean-field plots was

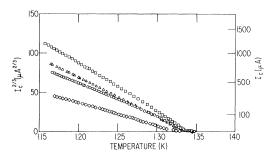


FIG. 3. $I_c^{2/3}(T)$ vs temperature for aluminum microbridges having $T_c=1.347$ K, thickness of 1200 Å, and widths of \Box , 2.45 μ m; Δ , 1.50 μ m; \bigcirc , 1.40 μ m; \bigcirc , 0.85 μ m.

less than the experimentally measured T_c . The solid line in Fig. 4 is the theoretical fit to Eq. (5). An extension of the curves to lower temperatures shows systematic deviations from the GL result below $T/T_c'=0.85$. This is to be expected since the theoretical result is valid only for temperatures near T_c , and should only provide an upper bound for critical current density at lower values of reduced temperature.

To check for nonuniform current distributions in the microbridges, the critical currents were measured with and without a superconducting ground plane. After first measuring the critical current as a function of temperature of the widest microbridge, a photoresist layer 1 μ m thick was spun on followed by deposition of 3000 Å of Pb. Subsequent critical current vs temperature measurements showed an increase of only 0.7% in the slope of this curve; the T_c of the bridge remained unchanged. Figure 5 shows these experimental results. The widths of all the other bridges measured were less than this particular microbridge, so that the error due to nonuniform current distributions is estimated to be less than 1%.

Figure 6 shows the experimentally determined $I_c(0)$ (obtained by extrapolating data obtained in the

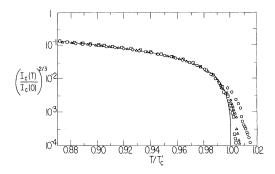


FIG. 4. Universal plots of $I_c^{2/3}(T)/I_c^{2/3}(0)$ vs T/T_c' for aluminum microbridges having $T_c=1.347$ K, thickness of 1200 Å, and widths of \Box , 2.45 μ m; Δ , 1.50 μ m; \Diamond , 0.85 μ m. The solid line is the GL result.

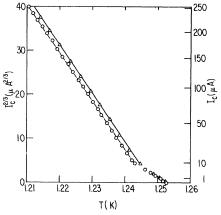


FIG. 5. $I_c^{2/3}(T)$ vs temperature for a $10 \times 2.15 \times 0.135$ - μm^3 bridge having a $T_c = 1.253$ K. The Δ graph is the microbridge with a 1- μ m layer of photoresist and a 3000-Å Pb ground plane evaporated over, and the O graph is the microbridge data without the ground plane.

mean-field regime near T_c to zero temperature) versus cross-sectional area for samples having three different T_c 's. Within experimental error the critical current scales with cross-sectional area so that the critical current density $J_c(0)$ can be obtained from the slope of each of these plots.

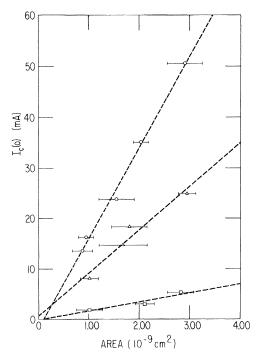


FIG. 6. Critical current extrapolated to zero temperature, $I_c(0)$, vs cross-sectional area for three transition temperatures; O, $T_c = 1.253$ K; Δ , 1.347 K; \Box , 2.355 K. The dashed lines are least-squares fit to the data.

| Thickness (Å) | <i>T_c</i> (K) | Resistivity ratio | $ ho_N \ (\mu\Omega\ 	ext{cm})$ | $	au_{ m resist.}$ (sec) | $J_c(0)$ (A/cm ²) | $	au_{ m crit.curr.} \ (m sec)$ |
|------------------|--------------------------|-------------------|---------------------------------|--------------------------|-------------------------------|----------------------------------|
| 6000 | 1.237 | 12.1 | 0.815 | 2.47×10^{-14} | 1.77×10^{7} | 4.44×10^{-14} |
| 1350 | 1.253 | 6.60 | 2.33 | 9.60×10^{-15} | 1.66×10^{7} | 3.68×10^{-14} |
| 1200 | 1.347 | 1.94 | 16.5 | 1.25×10^{-15} | 8.87×10^{6} | 8.53×10^{-15} |
| 320 | 1.439 | 1.77 | 22.8 | 8.58×10^{-16} | 1.01×10^{7} | 8.91×10^{-15} |
| 1280 | 2.355 | 1.03 | 953 | 2.21×10^{-17} | 1.75×10^6 | 6.28×10^{-17} |

TABLE I. Properties of aluminum weak links.

The impurity scattering is related to the experimentally determined $J_c(0)$ and T_c through Eq. (8). Using free-electron values, this can be written

$$\tau_{\text{crit. curr.}} = 2.63 \times 10^{-28} \sec J_c(0)^2 / T_c^3$$
 (9)

with $J_c(0)$ in A/cm² and T_c in K. τ obtained in this manner for our films decreases by three orders of magnitude in going from clean to dirty aluminum films as shown in Table I. The classical Drude-model formula¹ gives for the impurity scattering time obtained from 4.2-K resistivity measurements and free-electron value for N,

$$\tau_{\text{resist.}} = 1.95 \times 10^{-14} \text{ sec}/\rho_N$$
 (10)

with ρ_N in $\mu\Omega$ cm. Figure 7 summarizes all of our data. From this figure it can be seen that both methods give the same qualitative behavior of τ as a function of film granularity. Table I shows that

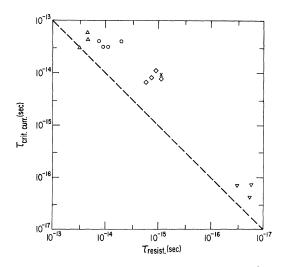


FIG. 7. $au_{\rm crit.\, curr.}$ (obtained from the critical current) vs $au_{\rm resist.}$ (from resistivity) for all 15 weak links from 5 different evaporations. The straight line represents $au_{\rm crit.\, curr.} = au_{\rm resist.}$. The average au calculated from the data for each evaporation are shown in Table I.

quantitatively they agree to within a factor of 10 in the worst case, and on average to within a factor of 6. The variation in mean free path with intrinsic film granularity shown in Table I is in reasonable agreement with the literature.⁹

It should be noted that the shortest scattering times $(\tau \sim 10^{-17} \text{ sec})$ correspond to mean-free paths of the order of less than 1 Å. In this regime, the concept of a mean-free path and the classical Boltzman formalism breaks down. It is interesting that the correlation between ρ , J_c , and T_c persists even for this nonphysical region. Therefore in this regime the proper way of looking at τ is as a parameter that describes the correlation of the normal-state and superconducting properties.

IV. SUMMARY

We have determined the magnitude and variation of the impurity scattering time of aluminum from separate measurements of the normal-state and the superconducting-state properties of aluminum microbridges. By adjusting the deposition parameters, we are able to make aluminum films having resistivities ranging from 1 to 1000 $\mu\Omega$ cm, resulting in films having impurity scattering times varying by three orders of magnitude. We have shown that from measurements of the critical current density and the transition temperature of aluminum in a microbridge configuration, and application of the Gorkov extension of the GL theory, the normal-state impurity scattering time can be obtained simply from the superconducting-state parameters $J_c(0)$ and T_c . The resulting τ 's agree quite well with those obtained from normal-state resistivity data and the classical Drude model formula.

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- *Also of the Department of Physics, Northwestern University, Evanston, Ill. 60201.
- ¹N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Holt, Rhinehart, and Winston, New York, 1975).
- ²L. P. Gorkov, Zh. Eksp. Teor. Fiz. <u>37</u>, 1407 (1959) [Sov. Phys. JETP <u>37</u>, 998 (1960)].
- ³M. Tinkham, Introduction to Superconductivity (McGraw-Hill, New York, 1975).
- ⁴N. R. Werthamer, in *Superconductivity*, edited by R. D. Parks (Marcel-Dekker, New York, 1969).
- ⁵Dektak Surface Profile Measuring System, Sloan Technology Company, Santa Barbara, Calif.
- ⁶C. M. Falco, W. H. Parker, S. E. Trullinger, and P. K. Hansma, Phys. Rev. B <u>10</u>, 1865 (1974).
- ⁷V. Ambegaokar and B. I. Halperin, Phys. Rev. Lett. <u>22</u>, 1364 (1969).
- ⁸C. M. Falco, T. R. Werner, and I. K. Schuller, Solid State Commun. <u>34</u>, 535 (1980).
- ⁹G. Deutscher, H. Fenichel, M. Gershenson, E. Grunbaum, and Z. Ovadyahu, J. Low Temp. <u>10</u>, 231 (1973).