

## Pair Conductivity above $T_c$ in Aluminum Films\*

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Pair conductivity, as reflected in the temperature dependence of the electrical resistance above the superconducting transition temperature, has been studied in aluminum films in the "two-dimensional" regime, viz.,  $d \lesssim \xi(T)$ , where  $d$  is the film thickness and  $\xi(T)$  is the superconducting coherence length. Normal-state resistivities were varied over five orders of magnitude, the highest-resistance films having highly disordered granular structures and the lowest-resistance films having mean free paths as large as 400 Å. The predicted temperature dependence of the pair conductivity was found to hold irrespective of sample parameters. However, the prediction that the excess conductivity should be independent of mean free path was found to not hold: Low-resistivity films exhibited an excess conductivity an order of magnitude larger than that predicted by Aslamazov and Larkin.

### I. INTRODUCTION

IN a recent letter<sup>1</sup> and paper<sup>2</sup> we reported a series of experiments dealing with the effect of thermodynamic fluctuations on the resistive transition of granular aluminum films in the temperature region near and below the transition temperature  $T_c$ . It was found that the resistance of these "two-dimensional" samples [i.e., samples with  $d \lesssim \xi(T)$ , where  $d$  is the film thickness and  $\xi(T)$  is the temperature-dependent coherence length] did not vanish abruptly, but instead, decayed exponentially with a decay rate linearly proportional to the normal resistance per square  $R_{\square}^n$ . The results of these experiments are in excellent agreement with a model recently proposed by Marčelja.<sup>3</sup>

As discussed in Ref. 2, the study of pair conductivity above  $T_c$  is plagued by various experimental and data handling problems not encountered in the experiments below  $T_c$ . We have now completed a study of pair conductivity above  $T_c$  in aluminum films, the results of which are presented below.

Aslamazov and Larkin<sup>4</sup> (AL) have presented the theory of pair conductivity above  $T_c$ , obtaining results which can also be derived from an application of the time-dependent Ginzburg-Landau theory.<sup>5,6</sup> A number of experimental studies of pair conductivity above  $T_c$  have been reported. In general, it is found that results obtained on extremely short-mean-free-path films prepared on cryogenic substrates ( $T \sim 4^\circ\text{K}$ ) are in reasonable accord with the AL theory.<sup>7</sup> Results obtained on extremely high-resistance granular aluminum films

are found also to be in satisfactory agreement with the AL predictions.<sup>8</sup> Anomalous results have been reported<sup>9</sup> for lead films prepared on nitrogen-temperature substrates (and annealed at room temperature). The present study differs from earlier ones in that a series of samples with values of mean free path varying over five orders of magnitude were studied. Moreover, it was possible, at least with some of the samples, to measure directly the value of the normal resistance per square,  $R_{\square}^n$ , thus eliminating the need for arbitrary parameters in the data analysis.

### II. THEORY

In order to compare the experimental data with the theory, it is necessary to calculate the excess conductivity  $\sigma'$ . This quantity is to be compared with the AL result:

$$\sigma' = (e^2/16\hbar d)(1/\epsilon), \quad (1)$$

where  $\epsilon = (T - T_c)/T_c$  and  $d$  is the film thickness. Since  $\sigma' = \sigma(T) - \sigma_n'$  it is apparent that, according to AL,

$$R_{\square}(T) = R_{\square}^n(1 + \epsilon/\tau_0)^{-1}, \quad (2)$$

where

$$\tau_0/R_{\square}^n = e^2/16\hbar = 1.52 \times 10^5 \Omega^{-1} \quad (3)$$

for any superconducting thin film, and  $R_{\square}$ , the resistance per square, is given by  $R_{\square} = R w/L$ , where  $w$  is the sample width and  $L$  is the sample length. It is convenient to rewrite Eq. (1) as

$$\epsilon/R_{\square}' = \tau_0/R_{\square}^n = e^2/16\hbar, \quad (4)$$

where

$$1/R_{\square}' = 1/R_{\square}(T) - 1/R_{\square}^n. \quad (5)$$

The criterion for two-dimensional behavior is that  $d \ll \xi(T)$ . If this condition is not met, it is necessary to include a correction  $G(T)$  to account for the nonzero

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<sup>1</sup> S. Marčelja, W. E. Masker, and R. D. Parks, Phys. Rev. Letters **22**, 124 (1969).

<sup>2</sup> W. E. Masker, S. Marčelja, and R. D. Parks, Phys. Rev. **188**, 745 (1969).

<sup>3</sup> S. Marčelja, Phys. Letters **28A**, 180 (1968); also Refs. 1 and 2.

<sup>4</sup> L. G. Aslamazov and A. I. Larkin, Fiz. Tverd. Tela **10**, 1104 (1968) [English transl.: Soviet Phys.—Solid State **10**, 875 (1968)].

<sup>5</sup> E. Abrahams and J. W. F. Woo, Phys. Letters **27A**, 117 (1968).

<sup>6</sup> A. Schmid, Z. Physik **215**, 210 (1968).

<sup>7</sup> See, for example, R. E. Glover, Phys. Letters **25A**, 542 (1967); D. C. Naugle and R. E. Glover, *ibid.* **28A**, 110 (1968); R. O. Smith, B. Serin, and E. Abrahams, *ibid.* **28A**, 224 (1968).

<sup>8</sup> M. Strongin, O. F. Kammerer, J. Crow, R. S. Thompson, and H. L. Fine, Phys. Rev. Letters **20**, 922 (1968).

<sup>9</sup> L. R. Testardi, W. A. Reed, P. C. Hohenberg, W. H. Haemmerle, and G. F. Brennert, Phys. Rev. **181**, 800 (1969).

thickness of the film, where  $G(T)$  is defined by<sup>10</sup>

$$G(T) = \sum_{n=0}^{\infty} \frac{1}{1+n^2(\pi\xi/d)^2} = \frac{1}{2} \left( 1 + \frac{d}{\xi} \coth \frac{d}{\xi} \right). \quad (6)$$

Including this correction, Eqs. (1) and (4) become

$$\sigma' = G(T) (e^2/16\hbar d) (1/\epsilon) \quad (7)$$

and

$$G(T)R' = R_{\square}^n \epsilon / \tau_0. \quad (8)$$

Thus, if the AL theory is correct, a plot of  $G(T)R'$  versus  $T$  should result in a straight line with intercept  $T_c$  and slope  $R_{\square}^n/\tau_0 T_c$ .

### III. SAMPLE PREPARATION

The granular aluminum films used in this study were prepared by evaporating aluminum in the presence of oxygen as described in Ref. 2. The clean films were prepared by evaporation from an aluminum-wetted tungsten wire source onto room-temperature glass substrates at a rate of about 100 Å/sec in a vacuum of  $1 \times 10^{-6}$  Torr. The resistivity of the clean samples was controlled by varying the sample thickness. The thickness measurements were made using a Tolansky (fringes of equal chromatic order) interferometer.

It was advantageous to work with specimens having a high resistance. Therefore, the films were trimmed into a rectangular zig-zag pattern having a large length to width ratio. This trimming operation also eliminated any tapered edges, which might have resulted if evaporation masks had been used to define geometries. The possibility of nonuniformity in the samples was reduced by confining the rectangular patterns (with typical lengths of 20 cm and widths of 0.5 mm) to an area of the film less than 1.5 cm<sup>2</sup> for most of the samples.

In order to eliminate the question of edge effects entirely, two cylindrical samples were prepared (samples *F* and *G*) by evaporating aluminum onto #56 enamel-covered copper wires which were rotating at a rate of over ten revolutions per second. The sample length was limited to about 0.5 cm in order to minimize nonuniformity. The thickness of the cylindrical films was calculated from Tolansky measurements made on a control slide, which had been placed alongside the rotating substrate. These measurements (for samples *F* and *G*) are subject to a larger uncertainty than that shown in Table I, because of the possible difference in the substrate-aluminum sticking coefficients between the sample and control slide substrates. This is not serious, however, since a knowledge of the film thickness was not needed in the determination of the quantity of interest,  $\tau_0/R_{\square}^n$ , for samples *F* and *G*.<sup>11</sup>

TABLE I. Sample parameters (defined in text).

Sample	$d$ (Å)	$R_{\square}^n$ (Ω/□)	$T_c$ (°K)	$\xi(0)$ (Å)	$10^5 \tau_0$	$\frac{\tau_0}{R_{\square}^n} / \left  \frac{\tau_0}{R_{\square}^n} \right _{AL}$
A	899 ± 44	0.111	1.223	2176	1.86	11.0
B	964 ± 11	0.115	1.220	2067	2.54	14.5
C	601 ± 50	0.369	1.275	1430	2.13	9.16
D	499 ± 13	0.387	1.275	1532	6.27	10.7
E	651 ± 32	0.507	1.320	1152	7.05	9.12
F	293 ± 60	0.906	1.338	1275	12.1	8.79
G	260 ± 26	0.922	1.280	1372	8.82	6.30
H	350 ± 50	1.86	1.380	802	20.6	7.28
I	136 ± 14	18.6	1.837	353	175	6.19
J	113 ± 23	25.8	2.050	311	235	5.99
K	248 ± 40	50.3	2.000	152	430	5.63
L	222 ± 43	141	2.090	93.9	1100	5.09
M	155 ± 32	203	2.188	91.4	1150	3.72
N	118 ± 20	350	2.235	79.2	1840	3.45
O	219 ± 22	722	2.266	40.9	2190	2.00
P	190 ± 40	1070	2.174	36.5	3230	1.99
Q	190 ± 40	1444	2.132	31.2	2940	1.34
R	130 ± 30	5106	1.875	22.1	4350	0.56 <sup>a</sup>

<sup>a</sup> Reference 18.

### IV. EXPERIMENTAL PROCEDURE

The samples were prepared for conventional four-probe resistance measurements by either applying silver paint contacts or soldering indium contacts to the glass substrate. The resistance of the samples (immersed in liquid helium) was measured in the temperature range 1.07–4.2°K by measuring the voltage across the sample, in the presence of a constant measuring current, with a Leeds and Northrup *K*-5 potentiometer. The measuring current was provided by a heavy-duty 12-V battery in series with a large metal foil resistor and was measured to be stable to within better than one part in  $10^5$ . The measuring current density, which ranged from 5 to 500 A/cm<sup>2</sup>, was chosen to be small enough to avoid current-dependent effects, but large enough to eliminate possible influence from noise currents generated by room-temperature circuitry.<sup>12</sup> The acceptable current range was determined by varying the current density by as much as a factor of 25 and observing only insignificant changes in the measured values of  $\tau_0$ . All measurements were made inside a copper-screened room to eliminate possible noise pickup from external sources. Temperatures were determined by a carbon-resistance thermometer placed in close proximity to the sample and calibrated in each run against the vapor pressure of the helium bath.

As is apparent from Eq. (5),  $R_{\square}^n$  is an extremely important quantity in the data analysis. Therefore, an effort was made to measure  $R_{\square}^n$  as accurately as possible. In the case of the cleaner films, the measurement of  $R_{\square}^n$  was straightforward. It was found that the pair conductivity in the cleaner samples (e.g.,

<sup>10</sup> H. Schmidt, Z. Physik **216**, 336 (1968).

<sup>11</sup> The film thickness enters Eq. (8) only through the quantity  $G(T)$  which can be approximated by unity for samples *F* and *G* (see discussion in Sec. V).

<sup>12</sup> The presence or absence of deleterious effects associated with noise currents generated by the room-temperature circuitry was established by inserting inductances in the sample leads and noting the change, if any, of the measured signal.

samples *A-E* in Table I) could be completely quenched (for all values of  $\epsilon$ ) by the application of a perpendicular magnetic field of approximately 50 G. Increasing the field from 50 to 350 G resulted in no measurable change (less than one part in  $10^5$ ) in the sample resistance, thereby demonstrating the unimportance of magneto-resistive effects. It was found that in the presence of a 350-G field there was no measurable (less than one part in  $10^5$ ) change in resistance when the samples were cooled from 2°K to a temperature lower than the zero-field transition temperature ( $\sim 1.2^\circ\text{K}$ ), which demonstrated that the temperature-dependent phonon-scattering contribution to the sample resistance was negligibly small. Therefore, it was possible to measure directly the value of  $R_{\square}^n$  for samples *A-E* (also for samples *F-H* by using somewhat larger magnetic fields).

For samples with resistances larger than 10  $\Omega/\text{square}$ , the fields required to quench the pair conductivity were inaccessibly large. In some cases, it was possible to estimate  $R_{\square}^n$  by measuring  $R$  as a function of  $T$  for temperatures between 2 and 20°K and then extrapolating a best fit to the high-temperature  $R$ -versus- $T$  data back to the region between 4.0°K and  $T_c$ . This analysis was complicated by the presence of "semiconducting" behavior (i.e., a small negative temperature coefficient of resistance far above  $T_c$ ).<sup>2</sup> For films having  $R_{\square}^n \lesssim 500 \Omega/\text{square}$ , it was possible to arrive at a fairly reliable estimate ( $\pm 0.1\%$ ) for  $R_{\square}^n(T)$  by the extrapolation procedure outlined above. For dirtier samples, the semiconducting behavior is stronger (see Table II of Ref. 2) and the procedure correspondingly less reliable. The estimated error in  $R_{\square}^n$  for 5000  $\Omega/\text{square}$  samples is about  $\pm 5\%$ . For this reason, the normal resistance of most of the high-resistivity films was treated as arbitrary parameters and chosen as that value of  $R_{\square}^n$  which gave the best fit to Eq. (1). It was found that to within 10% the value of  $\tau_0$  obtained from the measurements were independent of whether  $R_{\square}^n$  was measured or treated as an arbitrary parameter.

## V. EXPERIMENTAL RESULTS

For the purpose of checking Eq. (8), it was convenient to plot  $G(T)R_{\square}'$  versus  $T$ . For samples with  $R_{\square}^n \lesssim 100 \Omega/\text{square}$ ,  $G(T)$  is very nearly unity over the entire temperature region studied. In these cases, the correction factor  $G(T)$  can be omitted in the analysis with a resulting error in  $\sigma'(T)$  no greater than 2 or 3%. For samples with  $R_{\square}^n > 100 \Omega/\text{square}$ ,  $G(T)$  should be considered in the data analysis if the measurements extend past  $\epsilon \sim 0.2$ . When  $R_{\square}^n$  is greater than 1000  $\Omega/\text{square}$ , the effect of  $G(T)$  is of major consequence. This, unfortunately, means that the values of  $\tau_0$  measured for these very dirty samples are dependent upon the value of  $d$ , the film thickness, and  $l_{\text{eff}}$ , the effective mean free path, and that uncertainties in either of these quantities

will be reflected in an error in the value of  $\tau_0$ .<sup>13</sup> The values of  $\tau_0$  obtained for samples with  $R_{\square}^n \lesssim 1000 \Omega/\text{square}$  are unaffected by errors in either  $d$  or  $l_{\text{eff}}$ .

Figures 1(a)–1(d) show plots of  $G(T)R_{\square}'$  versus  $T$  for a clean planar sample, a clean cylindrical sample, a moderately dirty planar sample, and an extremely dirty planar sample, respectively. In Figs. 1(a) and 1(b),  $G(T)$  has been set equal to unity since the effect of this correction factor is negligible for these cleaner films. For both of these clean samples, the normal resistance  $R^n$  was measured with a magnetic field as described above. The effect of  $G(T)$  has been considered in Figs. 1(c) and 1(d), although, in the moderately dirty sample (sample *M*) the effect of setting  $G(T)$  equal to unity causes only a 20% error in the measured value of  $\tau_0$ . The value of  $R_{\square}^n$  for sample *M* [Fig. 1(c)] was extracted from a plot of  $R$  versus  $T$  between 2 and 20°K. Since the semiconducting behavior of this sample is small, the value of  $R_{\square}^n$  determined from the high-temperature data is relatively accurate.

The value of  $R_{\square}^n$  for the dirty sample [sample *O*, Fig. 1(d)] was treated as an arbitrary parameter and taken as that value which gave the best least-squares fit to Eq. (8). In this analysis, the value of  $T_c$  was also treated as an arbitrary parameter. It was found that the value of  $T_c$  determined in this way differed very little from the value of  $T_c$  determined by the criterion  $R \rightarrow 0$ . In the least-squares analysis, only data for  $R_{\square}(T)/R_{\square}^n \gtrsim 0.7$  were used in order to eliminate the critical region near  $T_c$ , where the AL theory is expected to break down. Since the quantity of interest,  $\tau_0$ , is measured from the slope of the  $R_{\square}'$ -versus- $T$  plot near  $T_c$  (i.e.,  $\epsilon \lesssim 0.2$ ), the value of  $\tau_0$  is happily insensitive to the choice of  $R_{\square}^n$  for very dirty samples and is the same to within  $\pm 15\%$ , whether  $R_{\square}^n$  is measured or treated as an arbitrary parameter.

As is apparent from the figures, the predicted linear temperature dependence [Eq. (8)] is exhibited by all of the samples in the range  $0 \leq \epsilon \lesssim 0.2$ . The cleaner samples show a gentle departure of  $R_{\square}'$  from strict linear dependence on temperature for larger values of  $\epsilon$ . The apparent strict linear dependence exhibited by sample *O*, even for large values of  $\epsilon$ , is misleading and merely reflects the fact that the linear dependence was forced by the data analysis for sample *O* described above.<sup>14</sup> For large values of  $\epsilon$  (viz.,  $0.5 \lesssim \epsilon \lesssim 1.0$ ),  $R_{\square}'$  is extremely sensitive to the choice of  $R_{\square}^n$ , especially in the cleaner samples, whereas for small values ( $0 \lesssim \epsilon \lesssim 0.2$ ), the sensitivity of  $R_{\square}'$  on  $R_{\square}^n$  is much smaller. For example, essentially perfect agreement with the temperature dependence of Eq. (1) can be forced upon the data for sample *E* between 1.3 and 2.0°K merely by adjusting  $R_{\square}^n$  slightly (five parts in  $10^5$ ) from its measured value.

<sup>13</sup> The quantity  $l_{\text{eff}}$  enters  $G$  through the standard relation,  $\xi(T) = 0.85[\xi(0)l_{\text{eff}}/\epsilon]^{1/2}$ ; see Eq. (6).

<sup>14</sup> This same procedure has been used in previous studies of pair conductivity in ultra-dirty films, e.g., see Refs. 7 and 8.

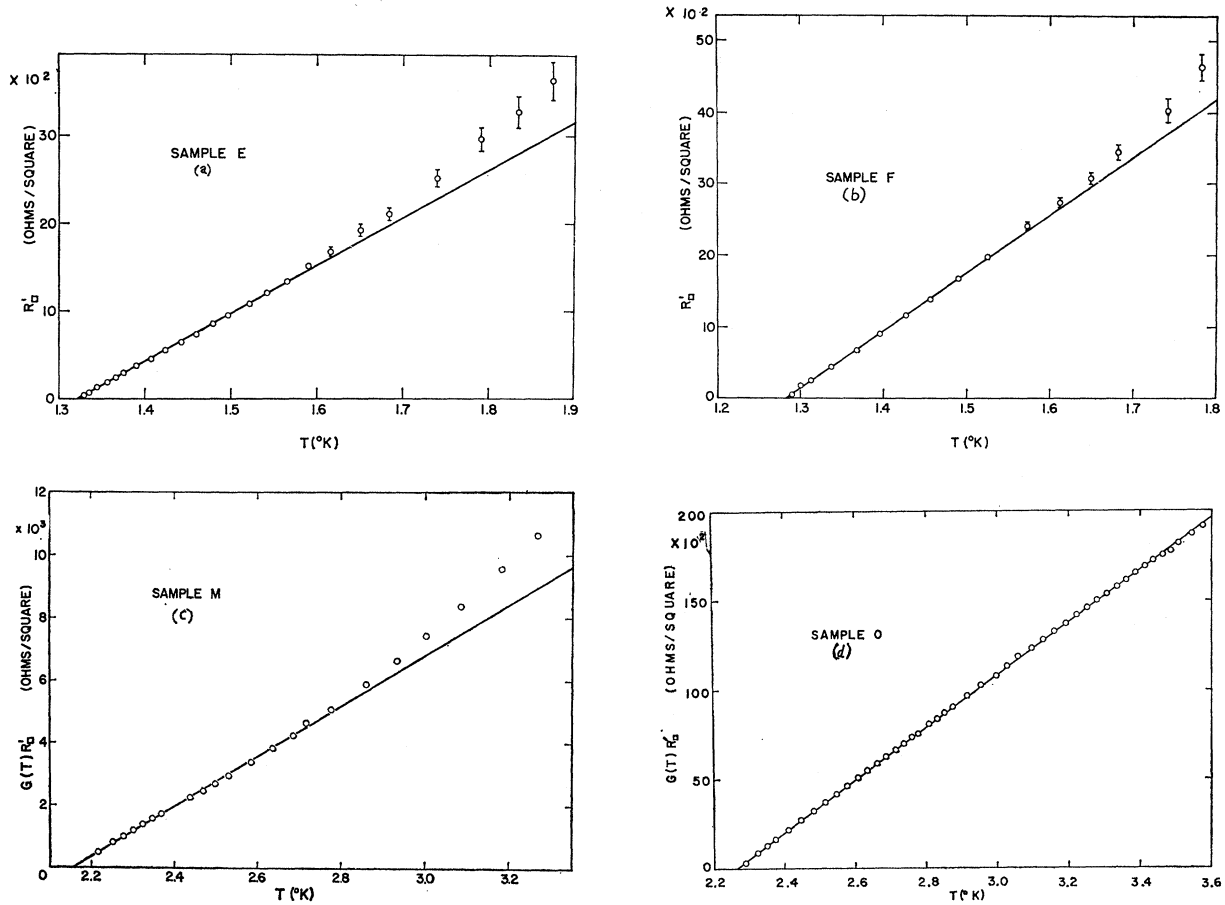


FIG. 1. (a) Temperature dependence of quantity  $R'_\square$ , which is defined by the relation,  $(1/R'_\square) = (1/R_\square) - (1/R_\square^n)$ , where  $R_\square$  is the measured resistance (per square) and  $R_\square^n$  is the normal resistance (per square), for sample E ( $R_\square^n = 0.507 \Omega/\text{square}$ ); (b) similar plot for (cylindrical) sample F ( $R_\square^n = 0.906 \Omega/\text{square}$ ); (c) temperature dependence of quantity  $G(T)R'_\square$  [where  $G(T)$  is defined in text] for sample M ( $R_\square^n = 203 \Omega/\text{square}$ ); similar plot for sample O ( $R_\square^n = 722 \Omega/\text{square}$ ).

The values of  $\tau_0$  were extracted from temperature plots, such as those in Fig. 1. These are tabulated, together with the sample parameters, in Table I. The values of  $\xi(0)$  listed in the table were calculated using the expression  $\xi(0) = 0.85(\xi_0 l_{\text{eff}})^{1/2}$  and the free-electron relation

$$\rho_n l_{\text{eff}} = \left[ \frac{2}{3} N(0) v_F e^2 \right]^{-1} = 0.40 \times 10^{-11} \Omega \text{ cm}^2 \quad (9)$$

as discussed in Ref. 2. Here  $\rho_n$  is the normal-state resistivity, and  $l_{\text{eff}}$  is the effective mean free path which has been shown by Abeles *et al.*<sup>15,16</sup> to provide a reasonably good description of transport processes in granular superconductors.<sup>17</sup> The values of  $T_c$  in the table are the values of the intercepts of the straight lines with the  $T$  axis in the  $G(T)R'_\square$ -versus- $T$  plots (Fig. 1). The higher values of  $T_c$  for samples ( $I$ - $R$ ) are not unusual for granular films and should be of no particular concern

in terms of the problem at hand, since it is assumed that the BCS law of corresponding states prevails.<sup>17</sup> The measured values of  $\tau_0/R_\square^n$  are compared with the value predicted by AL,  $(\tau_0/R_\square^n)_{\text{AL}}$ , in the last column of the table.

For illustrative purposes, the results  $\tau_0$  versus  $R_\square^n$  are plotted on a log-log grid in Fig. 2.<sup>18</sup> Included in the

<sup>18</sup> In the determination of the correction factor  $G$ , which was necessary to include in the analysis of results for the highest-resistivity films; Eq. (9) was used to relate  $l_{\text{eff}}$  to the measured value of  $\rho_n$ . In the case of sample R, for which the  $G$  correction is most important, the measured value of  $\tau_0/R_\square^n$ , which corresponds to incorporating the value  $\rho_n l_{\text{eff}} = 0.40 \times 10^{-11} (\Omega \text{ cm}^2)$  in the calculation of  $G$ , falls short of the AL line in Fig. 2. If, instead, the value  $\rho_n l_{\text{eff}} = 1.04 \times 10^{-11} (\Omega \text{ cm}^2)$ , which is suggested by the experimental results in Ref. 2, is used in the calculation  $G$ , the value of  $\tau_0/R_\square^n$  for sample R is larger (the triangle in Fig. 2). If the value,  $\rho_n l_{\text{eff}} = 1.6 \times 10^{-11} (\Omega \text{ cm}^2)$ , which is suggested by the critical-field study of a granular Al film with  $l_{\text{eff}} = 1.6 \text{ \AA}$  by Abeles *et al.* (Ref. 16), is used, this shifts the data point from below the AL line to slightly above it. Therefore, because of the uncertainty in our knowledge of  $l_{\text{eff}}$  in the superconducting state (see also the discussion in Ref. 2), we may say that the behavior of the most resistive samples studied (i.e.,  $R_\square^n \gtrsim 1000 \Omega/\text{square}$ ) is not in discord with the AL theory.

<sup>15</sup> B. Abeles, R. W. Cohen, and G. W. Cullin, Phys. Rev. Letters **17**, 632 (1966).

<sup>16</sup> B. Abeles, R. W. Cohen and R. W. Stowell, Phys. Rev. Letters **18**, 902 (1967).

<sup>17</sup> See discussion in Ref. 2.

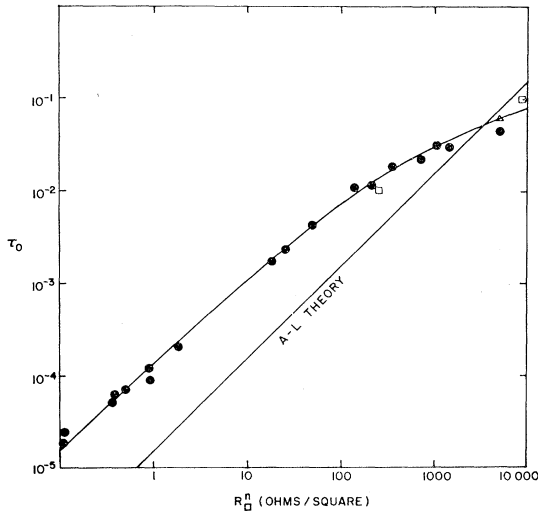


FIG. 2. Log-log plot showing dependence of measured parameter  $\tau_0$ , defined by  $\tau_0 = eR_{\square}^n/R_{\square}$ , on normal resistance (per square)  $R_{\square}^n$ . The straight line corresponds to the AL result,  $(\tau_0)_{AL} = e^2 R_{\square}^n / 16\hbar$ . See discussion in text and in Ref. 18.

figure are the results obtained Strongin *et al.*<sup>8</sup> (squares) on two short-mean-free-path Al samples (prepared by evaporating Al onto room-temperature substrates in a good vacuum and then exposing the film to oxygen). While the values of  $\tau_0/R_{\square}^n$  are in approximate agreement with the AL prediction in the case of the dirtiest films, they are over an order of magnitude larger than the predicted values for the cleanest films.

## VI. DISCUSSION

The following two features of the results presented above are curious: (1) the (anomalous) temperature variation of  $R_{\square}'$  for large values of  $T$  and, more important, (2) the (anomalous) variation of  $\tau_0/R_{\square}^n$  with  $R_{\square}^n$  (or mean free path). The first result can be easily disposed of, at least qualitatively. The simplest approach is to note that AL use no momentum cutoff in the conductivity integral which appears in the theory. A more reasonable approach would be to use an upper limit on the values of momentum  $q$  that enter the theory. For a superconductor, the largest momentum in the Ginzburg-Landau theory<sup>2</sup> is  $Q = 1/\xi(0) = 1/(\xi_0 l)^{1/2}$  which leads to the result

$$G(T)R_{\square}' = (R_{\square}^n/\tau_0)\epsilon(\epsilon+1) \quad (10)$$

instead of Eq. (8). By using Eq. (10) instead of Eq. (8), it is possible to obtain a better fit to the data in Figs. 1(a)–1(c). However, this would be a superficial approach since, for large values of  $\epsilon$ , the linearized mean-field theory itself fails, and more profound corrections must be made. The recent work of Gor'kov and Eliashberg<sup>19</sup> would seem to render even the latter

approach superfluous, their main thesis being that the linear approximation for the time-dependent Ginzburg-Landau equations (which was employed in both the AL and Abrahams-Woo theories) provides an inadequate description of a superconductor when frequencies other than extremely small ones are important (and most real problems fall in this category). It would thus seem that improvements in the existing theories are necessary before a meaningful discussion of this point can be given.

In regard to the anomalously large measured values of  $\tau_0$  for the cleaner aluminum films, it is perhaps relevant to mention some recent work of Thompson,<sup>20</sup> in which he calculates the excess conductivity by summing the AL term and a contribution proposed earlier by Maki.<sup>21</sup> Although the Maki term is divergent in two dimensions, Thompson claims that the presence of a weak pair-breaking interaction will remove the divergence and yield a simple expression for the conductivity, which is perhaps an order of magnitude larger than the AL value. A further increase in the strength of the pair-breaking perturbation will lead to a complete suppression of the extra term. A recent observation by Crow and co-workers<sup>22</sup> of a narrowing of the resistive transition in aluminum films (which corresponds to a decrease in  $\tau_0$ ) in an applied parallel magnetic field tends to support the Thompson model. However, it would be premature, we believe, to say that the question is closed; indeed, there seems to exist a general state of confusion regarding the treatment of the Maki term.<sup>23</sup> The diagram considered by Maki describes the correction to the normal electron conductivity due to the interaction of Cooper pairs with the sea of normal electrons. While it is intuitively obvious that this contribution cannot be divergent (the result<sup>21</sup> of first-order perturbation theory in two dimensions), it is possible that it can significantly affect the total conductivity, especially in cleaner samples (which are more apt to be free of pair-breaking effects<sup>24</sup>). The Maki term is unimportant below  $T_c$ , where the conductivity is dominated by the superfluid component, and is, therefore, of no concern in the experiments reported in Ref. 2.

One remaining point concerns the question of inhomogeneity broadening of the resistive transition. Although the possibility that sample inhomogeneity may enter prominently as an explanation of our results (in particular, the anomalously large values of  $\tau_0$ )

<sup>20</sup> R. A. Thompson, *Physica* (to be published).

<sup>21</sup> K. Maki, *Progr. Theoret. Phys. (Kyoto)* **40**, 193 (1968).

<sup>22</sup> J. E. Crow, R. S. Thompson, M. A. Klenin, and A. K. Bhatnager, *Phys. Rev. Letters* **24**, 371 (1970).

<sup>23</sup> E.g., open discussion in special session on the theory of fluctuations in superconductors at the International Conference on the Science of Superconductivity, Stanford, 1969 (unpublished).

<sup>24</sup> The small pair-breaking perturbation required to suppress the Maki term may be provided by localized magnetic moments in amorphous or granular films. Such moments could arise, in principle, because of quasilocalized electronic states in such systems which result either from extraneous impurities (e.g., oxygen) or local disorder.

<sup>19</sup> L. P. Gor'kov and G. M. Eliashberg, *Zh. Eksperim. i Teor. Fiz.* **54**, 612 (1968) [English transl.: *Soviet Phys.—JETP* **27**, 328 (1968)].

cannot be ruled out completely, plausibility arguments disfavor this explanation. For instance, it seems improbable indeed that inhomogeneity broadening would lead always to a linear dependence of  $R_{\square}'$  on  $T$  (for  $0 \lesssim \tau \lesssim 0.2$ ). Equally inexplicable, in terms of a sample inhomogeneity explanation, is the result that the data points for the 18 samples in the  $\tau_0$ -versus- $R_{\square}'$  plot (Fig. 2) all fall close to a smooth curve, in view of the fact that the samples vary widely in terms of the method of preparation and sample characteristics. In addition, our results on clean aluminum films have been corroborated in at least two other laboratories.<sup>24,25</sup> Finally, it has been found that samples with  $R_{\square}' \gtrsim 100 \Omega/\text{square}$  yield results which agree with Marčelja's extension of the theory below  $T_c$  but do not agree with the AL theory above  $T_c$ .

<sup>25</sup> M. A. Kleinin and M. A. Jensen (private communication).

*Note added in proof.* Thompson<sup>20</sup> predicts that the excess conductivity  $\sigma_a'$  arising from the Maki term in 2-dim films is given by

$$\sigma_a' = [2\tau_0/(\epsilon + \tau_0)] \ln[(\epsilon + \delta\tau)/\delta\tau],$$

where  $\delta\tau$  is the fractional decrease in  $T_c$  due to the presence of some pairbreaking perturbation. In the limit of small  $\epsilon$ ,  $\sigma_a'$  should become vanishingly small. An examination of the data close to  $T_c$  (e.g.,  $0 < \epsilon \lesssim 0.01$ ) for the clean samples used in the present study reveals that this does indeed happen (i.e.,  $\sigma' \approx \sigma_{AL}'$  in the limit of small  $\epsilon$ ).

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### Thermal-Conductivity Measurements of Gapless Behavior Produced by the Proximity Effect\*

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The thermal conductivities  $K_s$  and  $K_n$  in the superconducting and normal states have been measured for six In-Bi films of varying thickness with Mn layers on each side. The graphs of  $K_s/K_n$  against  $T$  show the characteristic linear variation of gapless systems near the transition temperature. In contrast to the expected behavior,  $K_s/K_n$  approaches a finite value at low temperatures. This is interpreted in terms of localized states near the  $n$ - $s$  boundary which cause gapless behavior at all temperatures.

#### I. INTRODUCTION

THE properties of a superconducting film are affected by the proximity of a normal metal in good contact with it, particularly if the normal metal is magnetic.<sup>1</sup> The most obvious effect is the reduction in the transition temperature. In addition both theoretical calculations and tunneling experiments have shown that there are profound changes in the density of states, such that there is no energy gap in the vicinity of the transition temperature. We have made thermal-conductivity experiments which confirm these features and which, in addition, show some unexpected behavior which may be interpreted in terms of localized states

near the boundary which cause a portion of the specimen to remain gapless (or have only a small gap) at all temperatures.<sup>2</sup>

For a description of our results and a comparison with the theory we consider two different temperature regions. Near the transition temperature the ratio of the superconducting and normal thermal conductivities ( $K_s/K_n$ ) varies linearly with temperature as for other gapless systems.<sup>3</sup> A quantitative comparison of theory and experiment requires that we take into account the spatial variation of the order parameter resulting from the proximity effect, and this is discussed in Secs. II A and V A.

At low temperatures the variation of  $K_s/K_n$  gives

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<sup>1</sup> For a review of the subject and a comprehensive list of references see G. Deutscher and P. G. de Gennes, in *Superconductivity*, edited by R. D. Parks (Marcel Dekker, Inc., New York, 1969), Vol. 2, Chap. 17.

<sup>2</sup> Two preliminary accounts of this work have been published: G. Deutscher, P. Lindenfeld, and R. D. McConnell, *Phys. Rev. Letters* **21**, 79 (1968); and in *Proceedings of the Eleventh International Conference on Low-Temperature Physics, St. Andrews, Scotland, 1968*, edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (University of St. Andrews Printing Dept., St. Andrews, Scotland, 1969), p. 993.

<sup>3</sup> K. Maki, Ref. 1, Chap. 18.