

## ELECTRICAL CONDUCTIVITY OF A TWO-DIMENSIONAL SUPERCONDUCTOR\*

S. Marčelja,† W. E. Masker, and R. D. Parks

Department of Physics and Astronomy, University of Rochester, Rochester, New York

(Received 16 December 1968)

The results of detailed experimental study of the temperature dependence of the electrical conductivity of a two-dimensional superconductor in the transition region are discussed in terms of a model which treats the critical region as well as the classical regions above and below the superconducting transition temperature  $T_c$ . The data taken below  $T_c$  suggest a means of distinguishing the contribution to the broadening of the resistive transition due to sample inhomogeneity from that due to intrinsic fluctuations.

Inexact off-diagonal long-range order in finite superconductors and the behavior of superconductors in the critical region are subjects of recent intensive interest.<sup>1</sup> Experimental studies to date have focused on the behavior of one-dimensional superconductors<sup>2</sup> [i.e., specimens with two dimensions smaller than the Ginzburg-Landau coherence length  $\xi(T)$ ] in the classical region below  $T_c$  and two-dimensional superconductors<sup>3</sup> in the classical region above  $T_c$ . In this Letter we present the results of an experimental study of the electrical conductivity of two-dimensional superconductors in the critical region as well as the classical regions above and below  $T_c$ , together with a model which describes the behavior throughout the entire transition region.

A consequence of inexact long-range order in one- or two-dimensional superconductors is that the conductivity, although extremely large, never becomes infinite in the condensed phase. This is due to the fact that below the nominal transition point there is no macroscopic occupation of the  $k=0$  level. There is, instead, a moderately large population of the few closely spaced, low- $k$  levels.<sup>4</sup> To attempt a quantitative description of this situation, we apply the model proposed by one of us (S.M.)<sup>5</sup> to the calculation of the conductivity of thin films. In the previous calculations valid above  $T_c$  the extra conductivity is proportional to the superfluid density and the lifetime of the superfluid excitations.<sup>6</sup> We extend the region of applicability of these models to the situation where the density of excitations is not small (i.e., the critical region) by including in the calculation the self-energy of the excitations due to their mutual interaction. The lifetime of an excitation is inversely proportional to its energy, and the constant of proportionality we choose in such a way as to give in the classical region above  $T_c$  the same value as the microscopic theory.<sup>7</sup> The general form for the extra conductivity due to the presence of superfluid in a two-dimen-

sional system is<sup>6</sup>

$$\sigma' = \frac{e^2}{4\pi^2 m d} \int_0^Q n(q) \tau(q) dq, \quad (1)$$

where  $n(q)$  is the  $q$  component of the superfluid density,  $\tau(q)$  is the lifetime of the superfluid excitations, and  $d$  is the film thickness. Using from Ref. 5 the expression derived for  $n(q)$  and the following one derived for  $\tau(q)$ ,

$$1/\tau(q) = (\alpha + \delta q^2 + \Sigma) 8k_B T \xi^2(0) / \pi \hbar \delta \quad (2)$$

[where  $\alpha = \delta(T - T_c) / T_c \xi^2(0)$  is the coefficient of the linear term in the Ginzburg-Landau (GL) theory,  $\Sigma$  is the self-energy of the excitations, and  $\delta = \hbar^2 / 2m$ ], we obtain

$$\sigma' = \frac{e^2 \delta^2}{16\pi \hbar d \xi^2(0)} \int_0^Q \frac{d^2 q}{(\alpha + \delta q^2 + \Sigma)^2}. \quad (3)$$

Equation (3) has the correct limiting form in the classical region above  $T_c$ , it remains continuous in the critical region, and below the critical region it reduces to (for  $Q = \infty$ , according to Aslamazov and Larkin)<sup>8</sup>

$$\sigma' = \frac{e^2}{16\pi \hbar d} \frac{\hbar^2}{2mk_B T \xi^2(0)} \exp\left(\frac{-4\pi\alpha\delta d}{\beta k_B T}\right), \quad (4)$$

where  $\beta$  is the coefficient of the tertiary term in the GL theory and has the value  $\pi\delta^2 / H_{c0}^2 [\xi(0)]^4$ ,  $H_{c0}$  being the thermodynamic critical field at  $T = 0^\circ\text{K}$ . Equation (4) is also the correct limiting form in the extremely dirty limit,  $\delta Q^2 \gg k_B T$ , even when one uses the finite cutoff,  $Q = 1/\xi(0)$ ,  $\xi(0)$  being the shortest length which occurs in the GL theory. For  $\delta Q^2 \ll k_B T$ , the factor  $\hbar^2 / 2m \xi^2(0) k_B T$  does not appear in Eq. (4). The exponential argument<sup>10</sup> is the same as that obtained by Kadanoff and Laramore,<sup>9</sup> who used a different approach. The disparity between the prefactor in Eq. (4) and the (temperature-dependent) one in

Ref. 9 reflects the use of the cutoff  $Q = 1/\xi(T)$  in the latter calculation.

The range of the off-diagonal long-range order  $r$  in the present model can be calculated to be (see, e.g., Ref. 4)

$$r = [\delta/(\alpha + \Sigma)]^{1/2}. \quad (5)$$

Equation (5) has the following asymptotic form below the critical region (for  $Q = \infty$ ):

$$r = \left(\frac{\delta}{k_B T}\right)^{1/2} \exp\left[-\frac{2\pi\delta\alpha d}{\beta k_B T}\right]; \quad (6)$$

i.e., the range of order increases exponentially with decreasing temperature. The disparity between this result and that obtained by Kadanoff and Laramore<sup>9</sup> reflects, again, the choice  $Q = 1/\xi(T)$  in Ref. 9.

The experimental portion of the present study consisted of measuring the "full" resistive transition curves (i.e., the resistance over several decades) of extremely short mean-free-path Al films. Aluminum was chosen because of the ease with which high resistivity Al films can be prepared, by using the method of Abeles, Cohen, and Collin,<sup>11</sup> wherein aluminum is evaporated in the presence of oxygen. Abeles, Cohen, and Cullin<sup>11</sup> have shown that such films are composed of crystallites smaller than 100 Å. In addition, it has been shown theoretically<sup>12</sup> that such films can be described by an effective mean free path  $l_{\text{eff}}$ , for  $p_F l_{\text{eff}} \gg 1$  ( $p_F$  being the Fermi momentum), even though the resistance is dominated by the tunneling barrier resistance between crystallites. Critical-magnetic-field measurements on granular Al films<sup>12</sup> have suggested that the effective-mean-free-path description is a good approximation even when  $p_F l_{\text{eff}} \sim 1$ . Therefore, when discussing the results below, we shall assume that such a description can be used for all of the films we have studied, and that the GL coherence length takes the form  $\xi(0) \sim (\xi_0 l_{\text{eff}})^{1/2}$ , where  $\xi_0$  is the value of  $\xi$  in the clean limit at  $T = 0^\circ\text{K}$  and  $l_{\text{eff}}$  is determined as discussed below.

The methods used to determine sample parameters and the preparation of the samples for conventional four-probe dc resistance measurements were straightforward and will be described, along with the details of the cryogenic setup, elsewhere.<sup>13</sup> The uncertainty in the relative temperature measurement was  $10^{-5}$  deg and the uncertainty in the relative dc resistance measurements was  $1:10^5$ . The dc-current density used in the resistance measurements was sufficiently

small (viz.,  $<20$  amp/cm<sup>2</sup>) to insure that the results were current independent.

The resistive transition for a very short mean-free-path 170-Å Al film (normal resistance per square  $R_{\square}^n = 5700 \Omega$ ,  $l_{\text{eff}} \sim 0.2 \text{ Å}$ ) is shown in Fig. 1. The quantity  $\ln \sigma_N/\sigma'$  is plotted against the absolute temperature,  $\sigma'$  being determined from the relation,  $\sigma = \sigma' + \sigma_N$ , where  $\sigma$  is the measured conductivity and  $\sigma_N$  the normal conductivity in the absence of superfluid fluctuations. The solid line in Fig. 1, which is a plot of Eq. (3), is seen to provide a reasonably good fit to the data over four orders of magnitude of the sample conductivity. The experiment parameters which enter Eq. (3) are  $\xi_0$ ,  $H_{c0}$ ,  $\sigma_N$ ,  $T_c$ , and  $l_{\text{eff}}$ . For  $\xi_0$  and  $H_{c0}$  we used the (nominal) values 16 000 Å and 100 G, respectively.<sup>14</sup> The normal conductivity  $\sigma_N$  was treated as an arbitrary parameter which gave the best fit to the high-temperature (Aslamazov-Larkin) limit of Eq. (3). The value

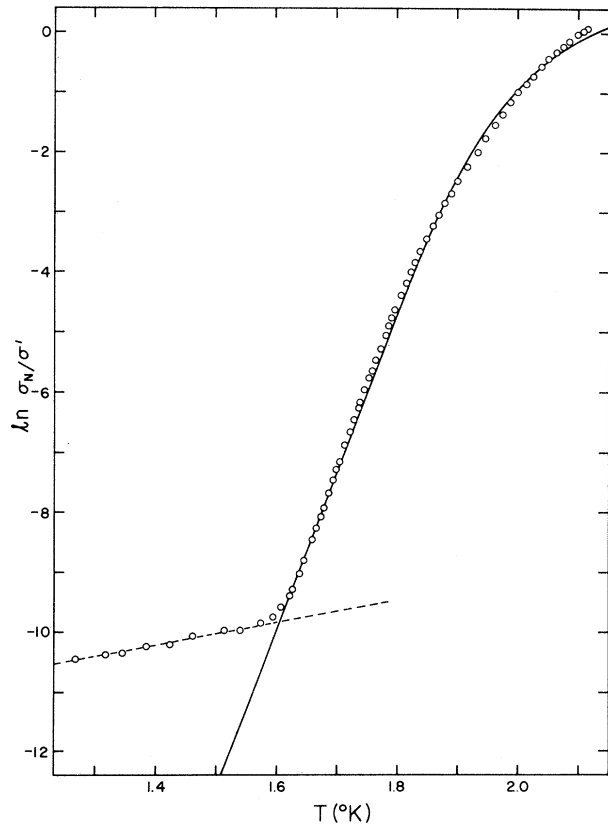


FIG. 1. Circles: resistive transition of Al film, whose parameters are given in text. The quantities  $\sigma_N$  and  $\sigma'$  are the normal conductivity and the excess conductivity due to the presence of superfluid excitation. Solid curve: plot of Eq. (3).

obtained in this way is only 5% larger than the value of  $\sigma$  at 4.2°K. For the purpose of testing the theory in the region of interest, viz.,  $\sigma'/\sigma_N \gtrsim 2$ , it is unimportant which of the two values of  $\sigma_N$  is chosen. The transition temperature  $T_C^{\text{nom}}$  was determined by extrapolating the data in the (linear) high-temperature portion of the curve of  $\sigma^{-1}$  vs  $T$ . Note that the quantity  $T_C$  which enters Eq. (3) differs from the nominal value  $T_C^{\text{nom}}$  according to the relation  $T_C = T_C^{\text{nom}}(1 + \epsilon_C)$ , where  $\epsilon_C$  (which has the value 0.009 for the sample under discussion) is approximately the width of the critical region.<sup>10,15</sup> This leaves as the only truly variable parameter the effective mean free path  $l_{\text{eff}}$ . The solid line in Fig. 1 corresponds to  $l_{\text{eff}} = 0.19 \text{ \AA}$ . It is of interest to compare this result with the value obtained from the normal resistivity, which we compute by using the result  $\rho l_{\text{eff}} = 1.6 \times 10^{-11} \Omega \text{ cm}^2$ , determined from skin-effect measurements made on pure Al.<sup>16</sup> This gives  $l_{\text{eff}} = 0.15 \text{ \AA}$ , which is, it seems, fortuitously close to the value obtained above, considering the questionable accuracy of determining the quantity  $l_{\text{eff}}$  from anomalous-skin-effect measurements and the questionable procedure of applying the effective-mean-free-path representation if  $p_F l_{\text{eff}} < 1$ . The upper part of the  $\sigma'^{-1}$  curve, viz.,  $-1 \lesssim \ln \sigma'_n / \sigma' \lesssim 5$ , which is independent of the mean free path, gave the value  $\epsilon_C / R_{\square}^n = 1.18 \times 10^{-5} \Omega^{-1}$ , which is 23% smaller than the predicted value.<sup>6</sup>

A major feature of the results in Fig. 1 is the sharp change in behavior of the experimental curve which occurs at  $T \sim 1.6^\circ\text{K}$ . The overall shape of the curve clearly suggests the presence of two resistance-broadening mechanisms. We identify the second of these which dominates at lower temperatures as arising from sample inhomogeneity. In the present study a variety of samples were studied with resistances in the range  $20 \lesssim R_{\square}^n \lesssim 6000 \Omega/\text{square}$ . The results in Fig. 1 are typical of those obtained with the high-resistance samples. As the resistance is decreased, the slope of the lower portion of the plot of  $\ln \sigma'^{-1}$  vs  $T$  increases relative to the slope of the upper portion, and the lower portion occupies an increasingly larger fraction of the resistive transition. For resistances of the order of  $R_{\square}^n = 100 \Omega/\text{square}$  the two regions become indistinguishable, suggesting that, for even smaller resistance samples, the entire resistance curve for  $T \lesssim T_C$  is dominated by sample inhomogeneity effects. For such samples the corresponding slope is much smaller (e.g., 1000

times smaller) than the theoretical value.

The discussion above suggests that experimental results obtained in the Aslamazov-Larkin (AL) regime ( $T > T_C$ ), particularly for relatively pure samples, might be dubious because of the presence of sample inhomogeneity effects. The slower variation of  $\sigma'^{-1}$  with temperature in the AL regime precludes the possibility of separating sample inhomogeneity effects from intrinsic fluctuation effects (which it was possible to do for  $T \ll T_C$  in the present study). Using pure Al films ( $l_{\text{eff}} \sim 1000 \text{ \AA}$ ), we have obtained excellent agreement with the predicted temperature dependence ( $\sigma'^{-1} \propto \epsilon$ ) in the AL regime, but the excess conductivity is larger by an order of magnitude than that predicted.<sup>13</sup> Whether this result is due to sample inhomogeneity is an open question at this time.

In summary, we have extended the existing mean field models of the resistance transition of a two-dimensional superconductor to include the critical region and temperatures lower than  $T_C$ . Certain results of this model are similar to those obtained recently by Kadanoff and Laramore.<sup>9</sup> Our experimental results on extremely dirty Al films are in good agreement with the model, but it appears that sample inhomogeneity obscures the observation of intrinsic fluctuation effects in pure Al films, at least for  $T \lesssim T_C$ . A more extensive report of this work will be published elsewhere.

We are grateful to L. P. Kadanoff and G. Laramore for sending us the manuscript of Ref. 9 before its publication.

Note added in proof. — It should be noted that the two-dimensional criterion  $d \lesssim \xi(T)$  is not strictly obeyed throughout the entire temperature interval in Fig. 1 [e.g.,  $d/\xi(1.6^\circ\text{K}) \sim 1.7$ ]. However, if Eq. (3) is corrected to account for this, the resulting change in  $\ln \sigma'/\sigma_N$  is negligibly small (e.g., the value of  $\ln \sigma'/\sigma_N$  at 1.6°K is changed by only a few percent). This correction leads, in fact, to a slightly better agreement between theory and experiment than that shown in Fig. 1.

\*Work supported in part by the U. S. Air Force Office of Scientific Research and the U. S. Army Research Office (Durham).

†On leave from the Institute of Physics, University of Zagreb, Zagreb, Yugoslavia.

<sup>1</sup>E.g., T. M. Rice, Phys. Rev. **140**, A1889 (1965); P. C. Hohenberg, Phys. Rev. **158**, 383 (1967); W. A. Little, Phys. Rev. **156**, 396 (1967).

<sup>2</sup>R. D. Parks and R. P. Groff, Phys. Rev. Letters **18**,

342 (1967); T. K. Hunt and J. E. Mercereau, Phys. Rev. Letters **18**, 551 (1967); W. Webb and R. Warburton, Phys. Rev. Letters **20**, 461 (1968); R. D. Parks, in Proceedings of the Conference on Fluctuations in Superconductors, Asilomar, California, 1968, edited by W. S. Goree and F. Chilton (Stanford Research Institute, Menlo Park, Calif., 1968), p. 141; W. Webb, *ibid.*, p. 159.

<sup>3</sup>R. E. Glover, Phys. Letters **25A**, 542 (1967); M. Strongin, O. F. Kammerer, J. E. Crow, R. S. Thompson, and H. L. Fine, Phys. Rev. Letters **20**, 922 (1968).

<sup>4</sup>D. A. Krueger, Phys. Rev. **172**, 211 (1968).

<sup>5</sup>S. Marčelja, Phys. Letters **28A**, 180 (1968).

<sup>6</sup>E. Abrahams and J. W. F. Woo, Phys. Letters **27A**, 117 (1968); A. Schmid, Z. Physik **215**, 210 (1968).

<sup>7</sup>E. Abrahams and T. Tsuneto, Phys. Rev. **152**, 416 (1966); A. Schmid, Phys. Condensed Matter **5**, 302 (1966).

<sup>8</sup>L. G. Aslamazov and A. I. Larkin, Fiz. Tverd. Tela **10**, 1104 (1968) [translation: Soviet Phys.—Solid State **10**, 875 (1968)], and Phys. Letters **26A**, 238 (1968).

<sup>9</sup>L. P. Kadanoff and G. Laramore, Phys. Rev. **175**, 579 (1968).

<sup>10</sup>Equation (4) may also be written in the more recognizable form,  $\sigma' \sim \exp(\epsilon/\epsilon_c)$ , where  $\epsilon = (T_c - T)/T_c$  and  $\epsilon_c$  is a fundamental parameter which has the value  $1.52 \times 10^{-4} R_{\square}^n$ ,  $R_{\square}^n$  being the normal resistance per square in ohms.

<sup>11</sup>B. Abeles, R. W. Cohen, and G. W. Cullin, Phys. Rev. Letters **17**, 632 (1966).

<sup>12</sup>B. Abeles, R. W. Cohen, and R. W. Stowell, Phys. Rev. Letters **18**, 902 (1967).

<sup>13</sup>W. E. Masker and R. D. Parks, to be published.

<sup>14</sup>The transition temperature of granular Al films is characteristically higher than the bulk value [e.g., see Ref. 12; also,  $T_c = 1.98^\circ\text{K}$  for sample A, whereas  $T_c(\text{bulk}) = 1.2^\circ\text{K}$ ]. This would lead to a decrease in the value of  $\xi_0$  and an increase in  $H_{c0}$ . However, the product  $H_{c0}^2 \xi_0^2$ , which appears in the argument of Eq. (3), should be relatively insensitive to the value of  $T_c$ , and the use of the bulk values for  $\xi_0$  and  $H_{c0}$  should lead to no serious error.

<sup>15</sup>The quantity  $T_c$ , which enters the theory, is defined as the temperature at which  $\alpha = 0$ .

<sup>16</sup>J. L. Olsen, Electron Transport in Metals (Interscience Publishers, Inc., New York, 1962), pp. 217-227.

## *g* SHIFT OF CONDUCTION ELECTRONS IN LITHIUM

A. W. Overhauser and A. M. de Graaf

Scientific Laboratory, Ford Motor Company, Dearborn, Michigan

(Received 9 September 1968)

The *g* shift of conduction electrons in lithium, determined recently by VanderVen, is about 30 times larger than a theoretically computed value. The general theory upon which the calculation was based is corrected. A revised theoretical estimate of  $\delta g$  agrees with experiment.

The *g* shift of conduction electrons in lithium has been measured accurately by VanderVen.<sup>1</sup> The experimental shift,  $\delta g = -6.1 \times 10^{-5}$ , differs substantially from the calculated value,  $\sim -2 \times 10^{-6}$ , of Bienenstock and Brooks.<sup>2</sup> Their calculation was based on the general theory of Yafet.<sup>3</sup> We have reinvestigated this theory and have found that it needs correction.

Energy bands in a crystal with inversion symmetry have spin degeneracy. It follows that one can define a *g* factor for each Bloch state  $\Psi_{n,\vec{k},s}(\vec{r})$ , where *n* is the band index,  $\vec{k}$  the wave vector, and *s* the spin direction,  $\uparrow$  or  $\downarrow$ . Each  $\Psi$  is a two-component Pauli function but is not, on account of spin-orbit coupling, an eigenfunction of  $\sigma_z$ . For each band *n*, the *g* factor  $g_n(\vec{k})$  is a function of  $\vec{k}$ . Sharp conduction-electron spin-resonance lines can occur only if the spin-lattice relaxation time *T* is much longer than the electronic scattering time  $\tau$ . The observed *g* is then a time-aver-

aged value, equal to the density-of-states average of  $g(\vec{k})$  over the Fermi surface.

The theory of the *g* shift is very complicated.<sup>3</sup> For convenience we divide  $\delta g(\vec{k})$  into two parts,

$$\delta g \equiv \delta g^P + \delta g^M. \quad (1)$$

$\delta g^M$  designates those terms in  $\delta g$  which would survive if the *g* shift of an atomic *S* state were calculated. We shall refer to  $\delta g^M$  as the Margenau term since Margenau was the first to compute *g* shifts of atomic states.<sup>4</sup>  $\delta g^M$  is finite even if the  $\vec{r}$  dependence of  $\Psi$  is independent of *s*.  $\delta g^P$  stands for all remaining terms.  $\delta g^M$  is a minor contribution to  $\delta g$  for all metals except Li. We have found that both  $\delta g^P$  and  $\delta g^M$  are given incorrectly by Yafet.<sup>3</sup> In this paper we shall derive the correct expression for  $\delta g^M$  and evaluate it for lithium.  $\delta g^P$  will be treated in a subsequent paper.