Electrical Conductivity of a Superconductor*

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A model is proposed which describes the dc electrical conductivity of a superconductor both above and below the classical critical point. The approach is to include in the calculation of superfluid density and the electrical conductivity, the effect of the fourth-order term of the Ginzburg-Landau theory, which represents the interaction between superfluid excitations. The resulting expression for the conductivity simplifies to the Aslamazov-Larkin result above T_c and yields, for "two-dimensional" samples, an exponential dependence of the superfluid conductivity on ΔT (= T_c – T) below T_c . Calculated results for the one- and three-dimensional cases are also given. Experimental studies of the electrical conductivity of "two-dimensional" Al films were made to test the model. Good agreement was obtained in the region below T_c , even when the sample resistance was followed over five decades and sample mean free paths were varied over two decades.

I. INTRODUCTION

INTIL rather recently it was generally accepted that it would be virtually impossible to observe thermodynamic fluctuations or critical-point effects in superconductors. This pessimism was generated in part by (correct) estimates of the extremely narrow temperature interval over which anomalous effects in equilibrium properties, such as specific heat, should be observable in bulk superconductors.¹ The outlook was brightened by Little's' discussion of the feasibility of observing the effect of thermodynamic fluctuations on the behavior of superconducting microgeometries. The first experiments'4 which provided at least qualitative evidence for fluctuation effects in superconductors were reported shortly thereafter. These experiments and subsequent related work⁵⁻⁷ focused on the critical current behavior of "one-dimensional" samples⁸ in the classical⁹ region below T_c . These experiments were particularly dificult because of sample preparation problems and the smallness of the measured effects.¹⁰

More recently, attention has shifted to the problem above T_c as a result of the paper by Glover¹¹ which

reported the sizable broadening for the resistive transition in thin amorphous Bi films, and the interpreting of these results by Ferrell and Schmidt¹² as arising from thermodynamic fluctuations. The theory was presented by Aslamazov and Larkin,¹³ by use of a microscopic approach. Later, Abrahams and Woo¹⁴ and Schmid¹⁵ obtained essentially the same result from the linearized time-dependent Ginzburg-Landau (GL) theory. Further time-dependent Ginzburg-Landau (GL) theory. Further
discussion has been given by Kadanoff and Laramore.¹⁶ The main result of these studies is the prediction that thermodynamic fluctuations of the order parameter above T_c lead to an excess conductivity σ' in the normal state, given by $\sigma' = \frac{\sigma_n \tau_0}{\epsilon}$, where $\tau_0 = \frac{e^2}{16d\hbar \sigma_n}$, d is the film thickness, σ_n is the normal conductivity, and $\epsilon = (T - T_c)/T_c$. This result is valid only in the twodimensional regime $[d \leq \xi(T)]$, in the dirty limit $(l \ll \xi_0$, where ξ_0 is the BCS coherence length and l the mean free path), and in the classical temperature region where the mean-field theory is expected to be a valid approximation. The last constraint requires that $\epsilon \ll 1$ and $\epsilon \gg 2\tau_0$, where $2\tau_0$ approximately defines the width of the critical region. '3 The one- and three-dimensional regimes, which are less accessible experimentally because of sample preparation difhculties in the first case and the smallness of the effect in the second, have also been treated. $13-15$

Glover's results on amorphous Bi films¹¹ and subse-Glover's results on amorphous Bi films¹¹ and subsequent results by Strongin *et al.*¹⁷ on Pb and Al films and quent results by Strongin *et al.*17 on Pb and Al films and
by Smith *et al.*18 on Pb films are all in reasonable accord with the predictions of the Aslamazov-Larkin theory.

The purpose of the present work was to extend both the theory and experiments to include the entire transi-

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tion region.¹⁹ Extending the theory from the classica region above T_c towards T_c requires the inclusion of the interaction between superfluid excitations, which becomes important as the density of excitations increases. Such interactions are represented by the fourth-order term of the GL free-energy functional. The model presented in the following section consists of including this term, in Hartree approximation, in the calculation of electrical conductivity. Detailed calculations for the two-dimensional case are presented in Sec. II, and the results for the one- and three-dimensional cases are given in Appendix A. Preliminary results on the calculation of specific heat within the spirit of the model are presented in Appendix B. The remainder of the paper reports new experimental results on Al films and com-

II. THEORY

pares these results with the predictions of the model.

For the reason of simplicity, we work with the Ginzburg-Landau formulation of the theory of superconductivity. In the transition region, this approach is equivalent to the BCS formulation and leads to the same results. The free-energy density for a given configuration of the order parameter is

$$
f(\psi,T) = a |\psi|^2 + \frac{1}{2}b |\psi|^4 + \delta |\nabla \psi|^2, \qquad (1)
$$

where the phenomenological constants $a, b,$ and δ are defined in Sec. V A. If we introduce $\psi = \sum_{k} \eta_k e^{i\mathbf{k} \cdot \mathbf{r}}$ and integrate over r, the free energy becomes (we consider unit volume)

$$
F(\psi,T) = \sum_{\mathbf{k}} (a + \delta k^2) \eta_{\mathbf{k}} \eta_{\mathbf{k}} \frac{\partial^2}{\partial b} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{Q}} \eta_{\mathbf{k} + \mathbf{Q}} \eta_{\mathbf{k}' - \mathbf{Q}} \eta_{\mathbf{k}} \eta_{\mathbf{k}'}.
$$
 (2)

In the quantized version of the theory η_k^{\dagger} , η_k may be regarded as creation and annihilation operators for superfluid excitations. The fourth-order term in its original form makes impossible the exact calculation of the partition function and correspondingly all other thermodynamic quantities. For any general state described by occupation numbers n_k the expectation value of the fourth-order term is

$$
\langle \cdots n_{\mathbf{k_2}} n_{\mathbf{k_1}} | \frac{1}{2} b \sum_{\mathbf{k}, \mathbf{k}', \mathbf{Q}} \eta_{\mathbf{k} + \mathbf{Q}}^{\dagger} \eta_{\mathbf{k}' - \mathbf{Q}}^{\dagger} \eta_{\mathbf{k}} \eta_{\mathbf{k}'} | n_{\mathbf{k}_1} n_{\mathbf{k}_2} \cdots \rangle
$$

= $b \sum_{\mathbf{k}} n_{\mathbf{k}} \sum_{\mathbf{k}'} n_{k'}.$ (3)

At this point, we invoke a Hartree-like approximation by replacing the sum over n_k by its average value,

$$
\sum_{\mathbf{k'}} n_{\mathbf{k'}} \sum_{\mathbf{k}} n_{\mathbf{k}} \approx n \sum_{\mathbf{k}} n_{\mathbf{k}} = \langle |\psi|^2 \rangle \sum_{\mathbf{k}} n_{\mathbf{k}}.
$$
 (4)

If we further assume the Bose character of the order-

parameter field,²⁰ n_k is given by

$$
n_{k} = \{ \exp[(a + \delta k^{2} + b \langle |\psi|^{2} \rangle) / k_{B} T] - 1 \}^{-1}, \qquad (5)
$$

and the average value of the superfluid density follows in a self-consistent way,

$$
\langle |\psi|^2 \rangle = \sum_{\mathbf{k}} \left\{ \exp\left[\left(a + \delta k^2 + b \langle |\psi|^2 \rangle \right) / k_B T \right] - 1 \right\}^{-1}.
$$
 (6)

Using the same approximation in the time-dependent GL equation, one obtains

$$
h\gamma(\partial/\partial t)\psi = a\psi + b\psi\langle |\psi|^2 \rangle + \delta\nabla^2\psi,
$$
 (7)

where γ $\left[\frac{-\pi \delta}{8k_BT \xi^2(0)} \right]$ is calculated in Ref. 21. Assuming the usual relaxation form for the solution $\psi_k = \psi_k(x) e^{-t/\tau_k}$, we obtain for the lifetime

$$
\tau_{\mathbf{k}} = \frac{\pi h \delta}{8k_B T \xi^2(0)} \frac{1}{a + \delta k^2 + b\langle |\psi|^2 \rangle}.
$$
 (8)

The superfluid conductivity $\sigma' = (e^2/m)\sum_{\mathbf{k}} n_{\mathbf{k}} \tau_{\mathbf{k}}$ is then

$$
\sigma' = \frac{\pi e^2 h \delta}{8m\xi^2(0)} \sum_{\mathbf{k}} \frac{1}{(a + \delta k^2 + b\langle |\psi|^2 \rangle)^2},\tag{9}
$$

together with Eq. (6) for $\langle |\psi|^2 \rangle$. In the equation for σ' , we have used the linearized form of Eq. (5) in order to simplify the algebra. For any real situation, this is an extremely good approximation. Equation (9) reduces to the Aslamazov-Larkin result¹³ in the classical region above T_c .

The above results are easily reproduced within the microscopic theory. However, a number of theoretical objections can be raised against the proposed approximation scheme. To begin, the Hartree approximation which we use in Eqs. (5) and (7) is a self-consistent treatment of the first order in the perturbation theory and will no longer be sufhcient at high superfluid densities. In three dimensions the approximation is not good close to the new (renormalized) critical temperature. In one and two dimensions, the present approximation gives no exact phase transition, but nevertheless, one expects the first-order theory to be a poor approximation when the Cooper pair density becomes quite large. This raises the more general question of the existence of an exact superconducting phase transition in one and two dimensions. At the moment, the theoretical situation on this equation is open²² and only a number of plausibility arguments can be invoked for either of the two possible answers. The last objection which we mention is the use of Bose statistics to describe a nonequilibrium system of Cooper pairs. As well as the previous assumptions, this approximation will become worse at higher pair densities.

¹⁹ Preliminary reports of this work are given in (a) S. Marčelja
Phys. Letters 28A, 180 (1968); (b) S. Marčelja, W. E. Masker, and
R. D. Parks, Phys. Rev. Letters 22, 124 (1969).

²⁰ For a discussion, see M. Revzen, Phys. Rev. $185, 337$ (1969).
²¹ A. Schmid, Physik Kondensierten Materie 5, 302 (1966).
²² See, e.g., W. D. Grobman, Phys. Rev. 182, 297 (1969),
and references therein.

In the remaining part of this paper we shall compare the presented model with the experimental results on thin films. The good agreement which is obtained supports our belief that the model has significantly extended the calculations of Aslamazov and Larkin,¹³ describing the electrical conductivity throughout the superconducting phase transition.

In the present work, we are interested in the solutions of the preceding equations applicable to thin films. Equations (6) and (9) become

$$
\langle |\psi|^2 \rangle = \frac{1}{d} \sum_{k_z} \int \frac{d^2 q}{(2\pi)^2}
$$

$$
\times \left[\exp\left(\frac{a + \delta(k^2 + k_z^2) + b\langle |\psi|^2 \rangle}{k_B T} \right) - 1 \right]^{-1}, \quad (10)
$$

$$
\sigma' = \frac{\pi e^{2\eta}\sigma}{8m\xi^2(0)d}
$$
\n
$$
\times \sum_{k_z} \int_0^Q \frac{d^2k}{[a+b\langle |\psi|^2 \rangle + \delta(k^2 + k_z^2)]^2}, \quad (11)
$$

where the film is assumed to be in the xy plane. Contrary to previous work, $13-15$ we have also introduced the cutoff momentum Q in the integrations. The justification for this is that the GL theory contains only first-order derivatives in the expression for free energy and is not valid for very high q values. For superconductors the highest value of q follows from the microscopic derivation of the GL theory, and is of order $1/\xi(0)$.²³ The introduction of the momentum cutoff $Q=1/\xi(0)$ in Eq. (11) does not affect the result close to T_c ($\epsilon \leq 0.1$), but acts to reduce the superfluid conductivity far above the transition. The approach, however, is superfluous for sufficiently large ϵ (i.e., $\epsilon \sim 1$), where the GL expansion itself is no longer a good approximation. After integration, (11) becomes

$$
\sigma' = \frac{e^2}{16hd} \frac{\delta}{\xi^2(0)} \sum_{k_z} \left(\frac{1}{a + b\langle |\psi|^2 \rangle + \delta k_z^2} - \frac{1}{a + b\langle |\psi|^2 \rangle + \delta/\xi^2(0) + \delta k_z^2} \right). \quad (12)
$$

The boundary condition is that the derivative of ψ should vanish at the surface. Both the real and imaginary parts of ψ can be expressed as Fourier series in $cos(\mathbf{k} \cdot \mathbf{r})$. This leads to the possible k_z values

$$
k_z = n\pi/d \,, \tag{13}
$$

where n is a positive integer (including zero).

The correction for finite film thickness takes a particularly simple form if we introduce the length [see,

e.g., Ref. $19(b)$],

$$
R = \left[\delta/(a+b\langle |\psi|^2 \rangle)\right]^{1/2},\tag{14}
$$

which describes the range of the order in the system. With decreasing temperature R becomes larger. To find the temperature dependence of R below T_c , we express Eq. (10) as

$$
a+b\langle |\psi|^2\rangle+\delta Q^2=k_B T[\ln(e^{\delta Q^2/k_BT}-e^{-4\pi\delta(|\psi|^2)/k_BT})
$$

-ln(1-e^{-4\pi\delta(|\psi|^2)/k_BT}). (15)

Below T_c , $\langle |\psi|^2 \rangle$ is large and approximately equal to $-a/b$. In dirty or moderately dirty metals $\delta Q^2/k_B T \gg 1$ and one obtains

$$
a+b\langle |\psi|^2 \rangle \approx k_B T e^{+4\pi \delta a/k_B T b}.\tag{16}
$$

The correction for film thickness is now similar to the one introduced earlier²⁴; the only difference is that the range of the order comes in place of the coherence length $\xi(T)$. If we introduce the notation

$$
G(x) = \sum_{n=0}^{\infty} \frac{1}{1 + n^2 x^2} = \frac{1}{2} \left(1 + \frac{\pi}{x} \coth^{-1} \frac{\pi}{x} \right),\tag{17}
$$

Eq. (12) may be written

$$
\sigma' = \frac{e^2}{16hd \xi^2(0)(a+b\langle |\psi|^2 \rangle)} \left[G\left(\frac{\pi R}{d}\right) - \frac{1}{1 + R^2/\xi^2(0)} \right]
$$

$$
\times G\left(\frac{\pi}{d\left[(a+b\langle |\psi|^2 \rangle)/\delta + 1/\xi^2(0) \right]^{1/2}} \right).
$$
(18)

For convenience we did not stop the summation over k_z at the maximum value Q. This introduces negligible error, as long as $d \sim \xi(T)$, since the first few terms in the sum are by far the most important; for $d\ll \xi(T)$ the term in brackets may be replaced by unity. It can be readily seen that the correction for finite film thickness will have negligible effect on the result below T_c . The reason for this is apparent: The contribution to σ' comes only from low k values and only the first term in the k_z sum is important as the range of order becomes much larger than the film thickness. In computing the value of σ' from Eqs. (10) and (16), we can also neglect the summation over k_z in Eq. (10). The correction to $\langle |\psi|^2 \rangle$ becomes important only for $T > T_c$, when $a\gg b\langle |\psi|^2 \rangle$, and then it does not change the result for σ' in Eq. (11) or Eq. (18).

Finally, we give the asymptotic form of the conductivity for $T \ll T_c$,

$$
\sigma' = \frac{e^2}{16hd} \frac{h^2}{2mk_B T \xi^2(0)} \exp\left(-\frac{4\pi\delta da}{bk_B T}\right).
$$
 (19)

This form is correct in the limit $\delta Q^2 \gg 1$; for $\delta Q^2 \ll 1$ the factor $\hbar^2/2mk_BT\xi^2(0)$ would be absent,

²³ P. G. de Gennes, Superconductivity of Metals and Alloys (W. A. Benjamin, Inc., New York, 1966), Chap. 7.

²⁴ H. Schmidt, Z. Physik 216, 336 (1968),

TABLE I. Sample parameters. The method used to determine ϵ_c and T_c is discussed in Sec. V A.

Sample	Thickness d (Å)	R_{\Box} ⁿ (Ω/sq)	$T_{\it e}$ (°K)	$10^3\epsilon_c$
	200	107	2.155	0.381
п	160	203	2.188	0.817
ш	120	350	2.325	1.32
IV	190	1070	2.174	4.04
v	190	1444	2.123	4.86
VT	170	4454	1.817	22.9
VII	170	4741	1.900	21.7
VIII	130	5106	1.775	15.4
IХ	170	6010	1.980	25.2

The analogs of Eqs. (6) , (9) , and (19) for the oneand three-dimensional cases are given in Appendix A.

III. SAMPLE PREPARATION

The system chosen to experimentally challenge the preceding model was that of thin, short mean-free-path Al films. This system is particularly attractive because of the ease and reproducibility with which films of widely varying resistivities can be made. The films were prepared according to the procedure of Abeles et al^{25} , wherein Al is evaporated in the presence of oxygen. Such films have been shown to be composed of very small crystallites with dimensions $\lesssim 100 \text{ Å}.^{25}$ very small crystallites with dimensions $\leq 100 \text{ Å}^{25}$ It has been shown theoretically by Parmenter²⁶ and by Abeles, Cohen, and Stowell²⁷ that hypothetical granular systems can be described by an effective mean free path, even though the resistance is dominated by the tunneling barrier resistance between grains (or crystallites). The effective transport mean free path is given by $l_{\text{eff}} = dt/(1-t)$, where d is the grain size and t the transmission coefficient between grains, and is related to the resistivity, as in ordinary metallic conduction, by (in the free-electron theory)

$$
(\rho l_{\rm eff})^{-1} = \frac{2}{3} N(0) v_F e^2 \,, \tag{20}
$$

where ρ is the resistivity, $N(0)$ is the density of states at the Fermi surface, and v_F is the Fermi velocity. While this result is strictly valid, within the approximations of the model, only for $p_F l_{\text{eff}} \gg 1$, where p_F is the momentum at the Fermi surface, critical magnetic field measurements made on granular Al films²⁷ imply that the effective mean-free-path representation is a good approximation even when $p_{F}l_{\text{eff}}$ 1. In particular, it was shown empirically that $\xi \approx (\xi_0 l_{\text{eff}})^{1/2}$ even when $p_F l_{\text{eff}} \sim 1$, a relation that is known to be valid when the mean free path is determined by impurity scattering. Therefore, when discussing the experimental results, we shall assume that all of the samples studied can be described by an effective mean free path given by $\rho_n l_{\text{eff}} = \text{const}$, and that the role of l_{eff} in terms of its effect on superconducting properties is the same whether its origin lies in impurity scattering or tunneling resistance. We admit that this method is open to challenge when $p_{F}l_{\text{eff}}$ < 1; yet, as will be shown, the results appear to be consistent with this assumption even for $p_{F}l_{\text{eff}}$ ~0.1.

In the present study, thin films were evaporated from aluminum-wetted tungsten wire sources onto roomtemperature glass substrates at oxygen pressures of 10^{-4} to 10^{-3} Torr. Evaporation rates were of the order of 100 Å/sec. The resistivity of the sample could be controlled by the amount of Al evaporated, the oxygen pressure, and the speed of evaporation. It was found that for thin films (150 Å or less) the resistance per square could be increased appreciably by merely exposing the sample to air for a period of days (or weeks).²⁸ Films prepared in this manner for the present study ranged in resistance from 100 to 6000 Ω /sq.

After deposition the film was cut into a rectangular zigzag pattern with a sharp tungsten needle mounted on a precision micromanipulator. This provided a sample with a large length to width ratio and correspondingly a large, easily measurable resistance. In general, the samples were constrained to areas smaller than 1 cm square in order to reduce the possibility of thickness variations due to evaporation source asymmetry or other causes. (For some of the "cleaner" samples, larger areas were required in order to provide enough sample resistance for accurate measurement at low currents.) An auxiliary benefit of the trimming operation was the presumed elimination of tapered edges, which always result from using evaporation masks to define geometries.

Film thicknesses which ranged 100—200 ^A were measured with a Tolansky interferometer to an accuracy of ± 20 Å. More accurate film thickness measurements were unnecessary since the film thickness does not enter the theory except through a small corrective term [the quantity G in Eq. (16)], this term being completely unimportant in the region of interest $T \lesssim T_c$.

IV. EXPERIMENTAL METHOD

The resistance of the Al samples was measured in the temperature range $4.2-1.07\,^{\circ}\text{K}$ by measuring the voltage across the sample in the presence of a constant measuring current. In the range 10 μ V–2 V, the voltage was measured to within 0.1 μ V with a Leeds and Northrup K-5 potentiometer. Lower voltages $(0.05-10 \mu V)$ were measured with a Keithley 1508 microvoltmeter. A heavy-duty 12-V automobile battery provided a measuring current which was constant to within one part in 10⁵. The measuring current densities ranged $2-20$ A/cm². It was found that within this range (typically $0.5-5 \mu A$, depending on the sample and temperature) the resist-

^{~&#}x27; B.Abeles, R. W. Cohen, and G. W. Cullin, Phys. Rev. Letters 17, 632 (1966). '

²⁶ R. H. Parmenter, Phys. Rev. 154, 353 (1967).
²⁷ B. Abeles, R. W. Cohen, and R. W. Stowell, Phys. Rev.
Letters 18, 902 (1967).

²⁸ This method of achieving higher resistances has also been used by Strongin and co-workers, e.g., Ref. 17, and M. Strongin, O. F. Kammerer, and A. Paskin, Phys. Rev. Letters 14, 949 (1965).

ance of the sample was ohmic and the result independent of measuring current; the measuring currents were, nonetheless, large enough to avoid the possible influence from noise currents generated by roomtemperature circuitry. All measurements were made inside a copper-screened room to eliminate possible noise problems from external sources.

The temperature of the samples, which were immersed in liquid helium, was determined from a carbon resistance thermometer placed in close proximity to the sample. The carbon thermometer was calibrated in each run against the vapor pressure of the helium bath. The temperature was controlled with a planar diaphragm manostat which was inserted in the pumping line to the Dewar. Regulated in this manner, the temperature was stable to approximately five parts in 10' in the normal helium range and one part in $10⁵$ in the superfluid range.

Since, as will be shown below, small magnetic fields were found to influence the sample behavior, it was necessary to provide magnetic shielding. The samples were mounted on a Formica insert and kept at least 10 in. away from the nearest metal support. The glass Dewar itself was placed inside a 4×10 -in. cylindrical magnetic shield which reduced the earth's field to less than 10^{-3} Oe. A copper solenoid magnet shrouded the Dewar inside the shield and could be used to provide small fields longitudinal to the Dewar's axis. The magnetic field inside the Dewar was measured before each run with a Hewlett Packard model 4283 milliammeter equipped with a Model 3529A magnetometer probe. The field longitudinal to the Dewar axis could be reduced to less than 2×10^{-4} Oe by applying small currents through the magnet.

The samples were mounted with the sample surface normal to the Dewar axis. Perpendicular magnetic fields could be applied with the solenoid in order to study the effect of field on the measured resistivity. Because of the small sample area, field uniformity was not a problem. The applied fields were kept deliberately small (less than 10 Oe) to avoid inducing a magnetic moment in the shield.

In order to fix the value of normal resistance and to examine the "semiconducting" behavior exhibited by the samples above T $_{c}$ (see Sec. V C), resistance measure ments were taken in the range $4.2-20$ °K for some of the samples. In these instances the samples were run inside a copper vacuum can immersed in liquid helium. An Advance wire heater was used to raise the sample temperature to 20'K. The temperature measurement in this range was provided by a carbon resistance thermometer which was calibrated in the liquid-helium range against the vapor pressure of helium and at approximately 20'K against the boiling point of liquid hydrogen. Temperatures between 4 and 20'K were determined using a standard-resistance-versus-temperature relation known to hold for the type of resistor used.

FIG. 1. Natural logarithm of resistance versus temperature for sample VI. Solid line: plot of Eq. (18) [which simplifies to Eq. (19) for $T \ll T_c$], with $R_{\Box}^{\eta} = 4454 \Omega/\text{sq}$ and $T_c = 1.817 \text{°K}$.

A check on this procedure was obtained by comparing the resistor with a germanium thermometer in the range 4—10'K.

7. EXPERIMENTAL RESULTS AND DISCUSSION

Nine Al samples, prepared in the manner discussed in Sec. III and having the parameters listed in Table I, were used in the study. As seen from the Table the T_c 's of the films varied and were always larger than the T_c of bulk Al. This is not a new result; the enhancement of T_c in such granular (or amorphous) films has been discussed extensively.²⁹ This provides no problem been discussed extensively.²⁹ This provides no probler in the present study, if we assume, as we shall below, that the BCS law of corresponding states holds for the hlms, which allows one to scale the relevant parameters according to T_c .

A. $T < T_c$

In order to test the model presented in Sec. II, in particular, Eqs. (10) and (11), the full resistive transitions of the samples were measured. The results for two extremely short mean-free-path samples are shown in Fig. 1 and Fig. 2, and those for a relatively clean sample in Fig. 3. The log plots tend to deemphasize the Aslamazov-Larkin (AL) region $(T \gg T_c)$ in favor of the region $T \leq T_c$, which is the region of particular interest in the present study.

The solid curves are plots of Eq. (11) [solved together with Eq. (10) . The thickness correction is

²⁹ See, e.g., Refs. 25, 26, 28; R. Hilsch, Non-Crystalline Solids, edited by V. D. Freschette (John Wiley & Sons, Inc., New York, 1958); M. Strongin, O. F. Kammerer, J. E. Crow, R. D. Parks, D. H. Douglass, Jr., and M. A. Jensen, Phys. Rev. Letters 21, 1320 (1968); J. W. Garland, K. H. Bennemann, and F. M.
Mueller, Phys. Rev. Letters 21, 1315 (1968).

FIG. 2. Resistive transition for sample VIII measured in "zero" magnetic field $\lfloor < 2 \times 10^{-4}$ Oe] and in applied perpendicular fields. Solid line: plot of Eq. (18) with $R_{\Box}^{n} = 5106$ Ω /sq and $T_c=0.1775$ °K.

negligibly small for $T \lesssim T_c$ and Eq. (17) is adequate to treat the classical region, corresponding to $\ln(R_{\Box}/R_{\Box}^{\quad n})$ ≤ -3 (R_{\Box} is the resistance per square and n refers to the normal state), which spans a sizable fraction of the measured curves. Equation (19) can be expressed in the more useful form

$$
\sigma' = \frac{e^2}{16hd} \frac{h^2}{2mk_BT\xi^2(0)} \exp\left(\frac{\epsilon T_c}{\epsilon_c T}\right),\tag{21}
$$

where $\epsilon_c = \epsilon b k_B T_c / 4\pi \delta da$.

Two adjustable parameters were used in fitting the theoretical curves in Figs. ¹—3 (and the corresponding curves for the other samples). These were T_c and ϵ_c , which were chosen to give the best agreement in the exponential region. Changing T_c has the effect of translating the entire curve horizontally, while changing ϵ_c has the effect of changing the slope of the curve in the exponential region. The T_c 's determined in this manner were in reasonable agreement with those determined by fitting the data to the theory in the Aslamazov-Larkin region above T_c (see Sec. V C).

The crucial test of the theory in the region $T < T_c$ below T_c reduces to that of demonstrating that the absolute magnitude of ϵ_c and the functional dependence of ϵ_c on R_{\Box}^n are consistent with the theoretical predictions [Eq. (21)]. For $\sigma' \gg \sigma_n$ we have $\sigma' \approx 1/\rho$, so that we may write

$$
\frac{d[\ln(R_{\Box}/R_{\Box}^{\ n})]}{dT} \approx \frac{T_c}{\epsilon_c T^2},\tag{22}
$$

which gives the slope of the resistance tail in the region
well below T_c . The measurement of this slope provides
a convenient test of the theory in the classical region
1969), p. 872,
1972, 872, a convenient test of the theory in the classical region

below T_c . The values of ϵ_c for the nine samples studied, which were extracted from theoretical fits to the data such as those shown in Figs. ¹—3, are listed in Table I and plotted as a function of R_{\Box} ⁿ in Fig. 4. The fact that the straight-line fit to the data has a slope of unity confirms the predicted linear dependence of ϵ_c on R_{\Box}^n . The equation of the line corresponds to ϵ_c = 0.40 \times 10⁻⁵ R_{\Box} ⁿ. The parameter ϵ_c can be expressed using the microscopic values for the constants in the GL free energy expansion.³⁰ Introducing $\delta = \hbar^2/2m$,

$$
a\!=\!\delta(T\!-\!T_c)/T_c\xi^2(0)\,,
$$

 $\xi(0) = 0.85(\xi_0 l_{\text{eff}})^{1/2}$, and $b = 1.02\hbar^2/Nl_{\text{eff}}^2m$, where N is the electron density, gives

$$
\epsilon_c = \frac{m v_F (1.04 \times 10^{-5})}{Ne^2 l_{\text{eff}} d}.
$$
 (23)

Finally, using $\lceil \text{Eq. } (20) \rceil$,

j

$$
R_{\Box}{}^{n} = \frac{m v_{F}}{N e^{2} l_{\text{eff}} d},\tag{24}
$$

one obtains $\epsilon_c = 1.04 \times 10^{-5} R_{\square}^{n}$, which is about 2.6 times the value determined from experiment. In view of this discrepancy, it should be noted that the expression for ϵ_c below T_c [Eq. (23)] contains a number of material constants, in contrast to the expression above T_c ($\sigma' = e^2/16hd$). We have eliminated the constants by use of Eq. (24), which might not be a good approximation for the amorphous Al films. A similar difficulty was encountered in the study by Abeles *et al*.,²⁷ in which the value of ρl_{eff} determined from upper critical magnetic field measurements on granular Al films was at least a

(22) FIG. 3. Resistive transition for sample II measured in zero field and in applied perpendicular fields. Solid line: plot of Eq. (18) with $R_{\Box}^{\text{v}} = 202.6 \Omega/\text{sq}$ and $T_c = 2.188^{\circ} \text{K}$.

factor of 2 greater than the value determined from Eq. $(24).^{31}$

In summarizing the above results, we note that the experimental results below T_c are in excellent agreement with the model in terms of confirming the predicted temperature dependence of σ' and the functional dependence of σ' on sample parameters. There remains the discrepancy in the absolute magnitude of ϵ_c (see Xote added in proof)

As seen in Figs. 1 and 3, the presence of a small perpendicular magnetic field leads to a severe distortion of the resistive tail. It was determined that the magnetoresistance is ohmic and, at temperatures for which $R_m \gg R$ (i.e., where the magnetoresistive tail is well separated from the zero-field tail in Fig. 1 or 3), linear with field. These properties of the magnetoresistance suggest that it results from current-induced resistance suggest that it results from current-induce
vortex flow.32 In addition, it was found that for current much larger than those used in the experiment a $V \propto I^2$ component appeared in the voltage (in the absence of field), which is expected in the flux flow state, when it is the field associated with the current, which produces the vortex state. The fact that flux flow is observed for perpendicular magnetic fields as small as 1 Oe and current densities as small as 10 A/cm^2 in such samples is not surprising. The field at which vortices enter (H_{c1}) is expected to be vanishingly small in such thin, short mean-free-path samples³³; the small depinning currents result presumably from the fact that the scale of the inhomogeneities which act as pinning sites is small compared to the size of the vortex core²⁵ $\lbrack \sim \xi(T)$, the size of the region over which the order parameter is depressed]. This is distinctly different than the situation in clean films or bulk materials, where the crystallite size is usually much larger than $\xi(T)$. A more complete report on flux-flow phenomena in such granular systems will be published elsewhere.³⁴ These observations are relevant to the present study only in that they document the need for canceling the earth's magnetic field in the experiments.

Sample inhomogeneity can also lead to anomalous tail broadening, which appears, in plots like those of Figs. 1 or 3, very much like the field-induced anomalies (e.g., Fig. 1 of Ref. 19b), except that it is relatively insensitive to magnetic field and does not vanish in the absence of field. While magnetoresistance (for $H \lesssim 2 \times 10^{-4}$ Oe) and sample inhomogeneity posed no

FIG. 4. Circles: R_{\Box} ⁿ versus the parameter ϵ_c for the nine samples whose parameters are listed in Table I; the values of ϵ_c are extracted from the best theoretical fits to the R -versus- T data [e.g., see Figs. 1-3]. Solid line: best straight-line fit to the data

problems, in the study of the samples reported in Table I, they represented serious obstacles in the study of cleaner samples (e.g., R_{\Box} ⁿ \lesssim 10 Ω /sq), because of the extremely rapid drop off of resistance in such samples. The resistive transitions in such samples were completely dominated by these deleterious effects.

B. $T \sim T_c$

While the agreement between the model and experiment is remarkably good in the region below T_c , it is clear from Figs. ¹—3 that the agreement is less good in the immediate vicinity of T_c . The mismatch between experiment and theory in the critical region seems to differ somewhat from sample to sample even though the sample parameters are quite similar (e.g., Figs. ¹ and 2), thus suggesting that at least part of the problem might lie in the experiment.

C. $T > T_c$ (Aslamazov-Larkin Regime)

The experimental investigation of the Al. regime poses special problems not encountered in the $T \leq T_c$ studies. The most serious of these is the extreme sensitivity of the excess conductivity on the normal resistance. A complicating aspect encountered in granular Al films (or amorphous films prepared at cryogenic temperatures) is the semiconducting behavior exhibited by such samples. For example, all of the samples used in the present study (Table I) exhibited small negative coefficients of resistance above T_c . This, which is not a coefficients of resistance above T_c . This, which is not new result,³⁵ arises presumably from the temperature dependent electron tunneling between Al grains sepadependent electron tunneling between Al grains sepa
rated by oxide.³⁶ This behavior is shown in Fig. 5, wher the temperature dependence up to 20'K is shown for

³¹ Note that there is an error in the value of l_{eff} used in Ref. 27 which reflects the same error in earlier sources (e.g., Ref. 13 of Ref. 27). The value of $\rho l_{\rm eff}$ for Al, based upon the measured values of the electronic specific-heat constant $[N]$. E. Phillips, Phys. Rev. 114, 676 (1959)], and the Fermi velocity from anomalous skin **114,** 676 (1959)], and the Fermi velocity from anomalous skin
effect measurements [E. Fawcett, in The Fermi Surface, edited by
W. A. Harrison and M. B. Webb (John Wiley & Sons., Inc.,
New York, 1961), p. 197], is ρl_{eff}

mately four times smaller than the value used in Ref. 27.

" For a review of flux flow in superconductors, see Y. B. Kim

and M. J. Stephen, in Superconductivity, edited by R. D. Parks

(Marcel Dekker, Inc., New York, 196

³⁵ See, e.g., O. F. Kammerer, D. H. Douglass, and M. Strongin, Bull. Am. Phys. Soc. 12, 417 (1967).
Bull. Am. Phys. Soc. 12, 417 (1967).
Thin Flm Physics, edited by R. Neidermayer and H. Mayer

⁽Vandenhoeck & Ruprecht, Göttingen, 1966), p. 579.

FIG. 5. Plot of resistance versus temperature for a representative sample, which reveals semiconducting behavior above T_c . Solid line: best fit to the data.

sample IV, which has an intermediate value of R_{\square} ⁿ. The dependence of this semiconductivity on the mean free path is illustrated in Table II, where the difference between $R_{\Box}^{\,n}(20^{\circ})$ and $R_{\Box}^{\,n}(10^{\circ})$ is shown for three samples which span a large range of R_0 ⁿ. Despite the semiconducting behavior, it is possible to choose a value of R_0 ⁿ which is correct to within 1 or 2%. This uncertainty is inconsequential in the theory for $T \lesssim T_c$ or in plots such as those in Figs. 1-3 which deemphasize the AL region, but is of crucial importance when determining σ' for $T \gg T_c$.

ien determining σ' for $T \gg T_c$.
Taking the lead of other investigators,^{11,17,18} we could force the data into the Curie-Weiss fit predicted by AL, i.e. ,

$$
\sigma' = \sigma - \sigma_n = \tau_0 T_c / (T - T_c) , \qquad (25)
$$

by treating both T_c and σ_n as arbitrary (constant) parameters, and then determine if $\tau_0^{\text{expt}} = \tau_0^{\text{AL},37}$ Doing parameters, and then determine if ${\tau}_0^{\rm expt} \! = \! {\tau}_0^{\rm AL, 37}$ Doing
this we obtain, as did Strongin and co-workers,¹⁶ reason able agreement with the AL theory for Al films with R_{\Box} ⁿ \gtrsim 3000 Ω /sq. For cleaner Al samples the excess conductivity, determined in this way, is larger than that predicted by AL, the discrepancy growing with increasing mean free path (e.g., for $R_{\Box}^{n} = 100 \Omega/\text{sq}$, τ_0^{expt} \sim 4 τ_0 ^{AL}). These results would seem to indicate (1) the breakdown of the AL theory for clean Al, or (2) sample inhomogeneity. The fact that our results on clean Al films have been corroborated in a second haboratory,³⁸ and that the behavior of the cleane samples used in the above study agrees with theory in the classical region below T_c , but not in the AL region, tends to support the former hypothesis. Since the $T\gg T_c$ study requires further experimental work because of the special problems associated with the AL region, as well as being not directly relevant to the main theme of this paper, a full report on it will be published

three representative samples.

Sample	$R_{\square}^n(T_c)$ (Ω/sq)	$R_{\Box}^{\,n}(10^{\circ}) - R_{\Box}^{\,n}(20^{\circ})$ $R_{\square}^n(10^{\circ})$
ТΤ	203	0.000452
TV	1070	0.0109
VIII	5106	0.0355

VI. SUMMARY

The theory of conductivity in superconductors has been extended from the classical region above T_c (AL region) to include the complete transition region. The approach was to calculate the superfluid density and excitation lifetime within the GL formalism, including the fourth-order term, which represents an interaction between superfluid excitations. The result for conductivity simplifies to the AL result for $T \gg T_c$ and gives strikingly diferent results for diferent dimensionalities in the classical region below T_c : for onedimension, $\sigma' \propto -\epsilon^3$; for two-dimensions, $\sigma' \propto e^{-\epsilon T_c/\epsilon_c T}$, where $\epsilon = (T - T_c)/T_c$ and ϵ_c is a sample-dependent parameter; and for infinite three-dimensional samples, $\sigma' = \infty$ for $T \leq T_c^*$ (thermodynamic fluctuations are manifested only as a small shift in T_c to a lower value T_c^*).

Experimental results on the electrical conductivity of short, mean-free-path two-dimensional Al films which span a large range of resistivities are in reasonable agreement with the model in the region below T_c . Experimental results on extremely short mean-free-path Al films in the region above T_c are in reasonable agreement with the AL (or present) theory; however, the excess conductivity exhibited by moderately clean Al films is larger than expected. Further work on long meanfree-path films is in progress.

Note added in proof. We have recently become aware of the size-effect work of I. Holwech and J. Jeppesen [Phil. Mag. 15, 217 (1967)] which leads to the value $\mu = 0.82 \times 10^{-11} \Omega$ cm² for Al at 4.2°K, which is approximately twice the value determined from a free-electron calculation. If the parameters a and b in Eq. (21) are evaluated in terms of the thermodynamic critical field and the (l-dependent) coherence length of bulk Al, instead of free-electron parameters, and if l is eliminated in favor of ρ (or R_{\Box}^{n}) by using the Holwech and Jeppesen result above, this leads to $\epsilon_c = 0.43 \times 10^{-5} R_{\Omega}$ ⁿ instead of $\epsilon_c = 1.04 \times 10^{-5} R_{\square}^{\prime\prime}$ and the factor of 2.6 discrepancy between experiment and theory discussed in Sec. V A is reduced to 1.1. This, rather than closing the question of a possible discrepancy, elucidates the fact that the value of ϵ_c is sensitive to material constants.

ACKNOWLEDGMENTS

It is a pleasure to thank Dr. R. Thompson for critically reducing the manuscript. We are grateful also

 ${\sf separately.} \over {\sf yr}$ It is possible to test the temperature dependence predicte by Aslamozov-Larkin only if T_c or R_\Box^n , or both, can be accuratel measured.

^{&#}x27;8 J. E. Crow and M. Strongin (private communication).

to Professor V. Ambegaokar, Professor R. Ferrell, Professor J. Langer, Professor K. Maki, and Professor A. Schmid for helpful discussions.

APPENDIX A: ELECTRICAL CONDUCTIVITY IN ONE AND THREE DIMENSIONS

In this Appendix we list the results for superfluid density and superfluid conductivity in one and three dimensions.³⁹ In one dimension the solutions corresponding to Eqs. (6) , (9) , and (17) are straightforward and read as follows:

$$
\langle |\psi|^2 \rangle = \frac{k_B T}{\pi \left[\delta(a+b \langle |\psi|^2 \rangle) \right]^{1/2}} \times \tan^{-1} \left(\frac{\delta Q^2}{a+b \langle |\psi|^2 \rangle} \right)^{1/2}, \quad \text{(A1)}
$$

$$
\sigma' = \frac{\pi e^{i\sigma}}{16hA\xi^2(0)} \frac{1}{(a+b\langle |\psi|^2 \rangle)^{3/2}},
$$
(A2)

or, below T_c ,

$$
\sigma' = \frac{\pi e^2 \delta^3 |a|^3}{2hA\xi^2(0)b^3k_B{}^3T^3} = \frac{1.71 \times 10^8}{A\xi^2(0)} \left(\frac{\epsilon L}{R^n}\right)^3, \quad \text{(A3)}
$$

where A is the cross-sectional area of the sample, L is the length, and $Rⁿ$ is the normal resistance in ohms. The results are insensitive to the value of the momentum cutoff and the form of the theory for high q values, and are expected to be a very good approximation. The resistance-producing mechanism described here is entirely different from and independent of the one proposed for one-dimensional systems by Langer and Ambegaokar,7 which leads to an exponential rather than cubic dependence of σ' on ϵ^{40}

The situation is more complicated in three dimensions. First we have to distinguish between two qualitatively different situations: an infinite 3-D system and a 3-D system of finite size. In the latter case, one may not replace the summation over k by integration. For the infinite 3-D case the first-order correction is insufficient close to T_c . Nevertheless, we give the results since they

represent improvement compared to linearized theory:

$$
\langle |\psi|^2 \rangle = \frac{k_B T}{2\pi^2 \delta} \left[Q - \left(\frac{a + b \langle |\psi|^2 \rangle}{\delta} \right)^{1/2} \times \tan^{-1} \left(\frac{\delta Q^2}{a + b \langle |\psi|^2 \rangle} \right)^{1/2} \right]. \quad (A4)
$$

This equation has a solution with $a+b\langle |\psi|^2 \rangle$ different from zero only if T is larger than the new critical temperature T_c^* . When $T = T_c^*$, $a+b\langle |\psi|^2 \rangle = 0$; this is the point of Bose-Einstein condensation and below this point the superfluid density has the same form as in the mean-field theory without fluctuations. The shift in the critical point is given by

$$
\Delta T_c = T_c - T_c^* = T_c \frac{k_B T b \xi(0)}{4\pi^2 \delta}.
$$
 (A5)

The conductivity is infinite below the new critical point and above the critical point is given by

$$
\sigma' = \frac{e^2}{32\xi^2(0)} \frac{1}{\left[2m(a+b\langle|\psi|^2\rangle)\right]^{1/2}}.\tag{A6}
$$

Experimentally, this situation is indistinguishable from the result of the linearized theory.¹³⁻¹⁵

In the case when one, two, or three dimensions are finite, we must retain the summation over k . In this case the superfluid conductivity below the transition is no longer perfect. For example, if the sample is a cube of side \bar{L} , we have

$$
\langle |\psi|^2 \rangle = \frac{k_B T}{L^3} \sum_{\mathbf{k}} \frac{1}{a + \delta k^2 + b \langle |\psi|^2 \rangle},\tag{A7}
$$

$$
\sigma' = \frac{\pi e^2 h \delta}{8m\xi^2(0)L^3} \sum_{\mathbf{k}} \frac{1}{\left[a + \delta k^2 + b\langle |\psi|^2 \rangle\right]^2}, \quad (A8)
$$

and for $-\epsilon \ll \epsilon_c$ (retaining only the first term in the summation),

$$
\langle |\psi|^2 \rangle = \frac{k_B T}{L^3 (a + b \langle |\psi|^2 \rangle)} \tag{A9}
$$

$$
\langle |\psi|^2 \rangle \!=\! -a/b \!-\! k_B T/L^3 a \,, \eqno({\rm A10})
$$

and

 α r

 α r

$$
\sigma' = \frac{\pi e^2 h \delta}{8m\xi^2(0)L^3} \frac{1}{(a+b\langle|\psi|^2\rangle)^2}
$$
 (A11)

$$
\sigma' = \frac{\pi e^2 h \delta}{8m\xi^2(0)} \frac{a^2 L^3}{b^2 k_B^2 T^2}.
$$
 (A12)

In the dirty limit the last equation reduces to

 $\pi \rho^2 h \delta$

$$
\sigma' = 0.72 \left(m^2 L^3 k_B T v_F / \hbar^3 \right) \sigma_n. \tag{A13}
$$

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³⁹ Throughout this paper the dimensionality of the quantities $\langle |\psi|^2 \rangle$ and b changes according to the number of dimensions of the system. The 3-D numerical value for b must be divided by the sample thickness for the two-dimensional case or the sample cross section in the case of one dimension.

⁴⁰ We believe that for temperatures close to T_c where the longwe other is incomplete, the present model is applicable. It is
possible that at sufficiently low temperatures long-range order prevails and the mechanism proposed by Langer and Ambegaokar is the dominant loss mechanism in one dimension. However, whether exact long-range order appears at any temperature in one- or two-dimensional systems is an open question, and one that is intimately related to the discussion of the validity of the approximations used in the present model, given in Sec. II.

As expected, the superfluid conductivity i large (larger than the upper limit determined from persistent current experiments⁴¹).

APPENDIX B: SPECIFIC HEAT AT THE SUPERCONDUCTING TRANSITION

The calculation of specific heat at the superconducthas been the subject of several investiga tions.^{1,42,43} All of these calculations a b in the subject of several investigation has been the subject of several investiga-
 $\frac{42.43}{41}$ All of these calculations are essentially

bation-theoretic expansions, divergent at the critical point. Correspondingly, the r in the region where the correction to the specific heat is Il compared to the discontinuity which is the result of mean-field theories

The self-consistent approximation from Sec. II can ied to this problem. The free energy of a super fluid excitation

$$
\mathcal{E}(\mathbf{k}) = a + \delta k^2 + b n , \qquad (B1)
$$

where $n = \langle |\psi|^2 \rangle$ describes the free energy required to add an excitation in the presence of superfluid. To obtain the free energy of condensation for the system in a $e,$ it is necessary to integrate from zero super ound density to its final value

$$
F = \int_0^n (a+bn)dn + \delta \sum_{\mathbf{k}} k^2 n_{\mathbf{k}}.
$$
 (B2)

The central result is then the partition function

$$
Z = \int \pi d n_{\mathbf{k}} \exp\left[-\frac{1}{k_B T} \sum_{\mathbf{k}} \left(a + \delta k^2 + \frac{b}{2} \sum_{\mathbf{k}'} n_{\mathbf{k}'}\right) n_{\mathbf{k}}\right].
$$
 (B3)

We have been unable to calculate (B3) exactly. In order to obtain the *complete* free energy and the specific heat, a number of approximations may be considered. In three dimensions, the present approximation (Sec. II) breaks down close to T_c . The one- and twocases are more amenable to approximation schemes, since in these cases there is no singular phase transition point and all the quantities are continuous and finite throughout the transition region. One of the

FIG. 6. Expected specific-heat anomaly [from Eq. $(B4)$] for 500 Å Al film with electronic mean free path of 500 Å. The plot onds to the total specifi Dashed line: classical th

possible approximations for the complete free energy is the following:

(B2)
$$
F = \sum_{k} \{ [a + \frac{1}{2}bn(T) + \delta k^{2}]n_{k}(T) \}.
$$
 (B4)

Since the interaction of the excitations enters only through the average value of the superfluid density, the free energy has been approximated in (B4) as a product of the free energy per excitation and the average number of excitations. Once the approximation for the complete free energy is given, the calculation of the specific heat is straightforward, but somewhat lengthered

fic heat has an extremely narrow However, as noted above, th ak at the new (shifted) transiti iterally. In the case of two di where one can apply the approximation in $(B4)$ with more confidence, the specific-heat peak (which rises well above the classical value at T_c) is round to lower temperatures as shown in Fig. 6. Qualitatively, this effect has been observed in near-monolayer helium films.⁴⁵ films.⁴⁵

⁴¹ See, e.g., J. File and R. G. Mills, Phys. Rev. Letters 10,

² A. P. Levanyik, Fiz. Tverd. Tela 5, 1776 (1963) [Englis 93 (1963).

² A. P. Levanyik, Fiz. Tverd. Tela 5, 1776 (1963) [English

transl.: Soviet Phys.—Solid State 5, 1294 (1964)]; V. G. Vaks,

A. I. Larkin, and S. A. Pikin, Zh. Eksperim. i Teor. Fiz. 51, 361

y of Maryland Technical Report No. 932, 1969 (unpublished).

⁴⁴ The details of the calculation will be described elsewhere se, ^p sica 15, 860 {1949).