Fluctuation Rounding of the Resistive Superconducting Transition in Thin Lead Films

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The effect of fluctuations on the resistive transition to superconductivity has been studied in thin (100–200 Å) Pb films. Although the temperature dependence of the fluctuation contribution is given by $(T-T_c)^{-1}$, and the width of the transition increases with increasing film resistance as predicted by Aslamazov and Larkin, this width is found to be smaller than the theoretical value by factors of 2–14. The transition from two- to three-dimensional behavior, expected theoretically in one of the samples studied, was not observed. A similar failure is found on analyzing published data for Al and (possibly for) amorphous Bi films. The various approximations used to derive the theory, and their effect on the analysis of the data (which has been carried out with no fitted parameters), are discussed in detail. The quantitative applicability of the Aslamazov-Larkin theory is considered in the light of its discrepancies with experiment. However, no explanation is found consistent with all published data. For one sample, measurements were performed in a magnetic field; a discussion of additional broadening mechanisms in a magnetic field is presented.

I. INTRODUCTION

THE observation of intrinsic fluctuation effects on the resistive transition to superconductivity of amorphous bismuth films has recently been reported by Glover.[1] Fluctuation effects, which are normally unobservable[2] in clean bulk samples (except for $|T-T_c|/T_c \lesssim 10^{-14}$), extend over a measurable temperature interval in this case, because of the short mean free path in this disordered structure and because of the two-dimensional nature of the sample. Glover found that above T_c, the fractional change in resistance R from the normal-state value R_N was given by

$$(R_N-R)/R_N=\tau_0/\tau,$$

where $\tau\equiv(T-T_c)/T_c$ and the width factor τ_0 is a constant. Earlier, and independently, Aslamazov and Larkin[3] (AL) had treated the fluctuation effects theoretically in the "mean-field" region, $(R_N-R)/R_N\ll 1$, and obtained the observed temperature dependence. They also found the remarkably simple result that

$$\tau_0=(e^2/16\hbar)R_N{}^{\square}\equiv\tau_0{}^{\mathrm{AL}},$$

where $R_N{}^{\square}$ is the normal-state "sheet resistance" of the film. The theoretical value of τ_0 was in excellent agreement with Glover's results. Subsequent measurements on Al[4] and Pb[5] films confirmed the τ^{-1} dependence and, with one possible exception,[4] also the theoretical value of τ_0.

We present in this paper an experimental study of the rounding of the resistive superconducting transition in thin lead films which fails to confirm the theoretical value of τ_0. Our data show the τ^{-1} temperature dependence but with coefficients τ_0 which are *less* than the theoretical value (by factors of 2–14), and not proportional to R_N. Our analysis shows, further, that the change in temperature dependence, due to finite thickness corrections, which theoretically should occur in some of the present and previously published data,[1,4] is not observed.

In Sec. II we discuss the theory and the approximations contained therein. Following a description of the experimental apparatus and techniques in Sec. III, we present, in Sec. IV, our results. In Sec. V we discuss several possible sources of the discrepancy, both experimental and theoretical. These include nonuniform current flow, incorrect analysis of the data, sample inhomogeneities, finite thickness corrections, phonon effects, and corrections to the theory of superconductivity due to the highly disordered nature of our samples. None of these effects appears to explain all experimental data. We also relate our results to those published by Glover,[1] Strongin *et al.*,[4] Smith *et al.*,[5] and Masker and Parks.[6]

II. THEORY

Aslamazov and Larkin[3] have calculated the electrical conductivity above T_c due to fluctuation pairing of electrons for two- and three-dimensional samples. We have evaluated their expression for arbitrary thickness d, using the boundary condition that the derivative of the order parameter should vanish at the film edge. We

[1] R. E. Glover, Phys. Letters **25A**, 542 (1967). More recent experimental work confirming the earlier investigation was reported by R. E. Glover, in *Proceedings of the Eleventh International Conference on Low-Temperature Physics*, edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (University of St. Andrews Printing Department, St. Andrews, Scotland, 1969), Vol. II, p. 793; D. C. Naugle and R. E. Glover, Phys. Letters **28A**, 110 (1968).

[2] See, for instance, P. C. Hohenberg, in *Proceedings of the Eleventh International Conference on Low-Temperature Physics*, edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (University of St. Andrews Printing Department, St. Andrews, Scotland, 1969), Vol. I, p. 33.

[3] L. G. Aslamazov and A. I. Larkin, Fiz. Tverd. Tela **10**, 1104 (1968) [English transl: Soviet Phys.—Solid State **10**, 875 (1968)]. A preliminary account of this work was published in Phys. Letters **26A**, 238 (1968).

[4] M. Strongin, O. F. Kammerer, J. Crow, R. S. Thompson, and H. L. Fine, Phys. Rev. Letters **20**, 922 (1968).

[5] R. O. Smith, B. Serin, and E. Abrahams, Phys. Letters **28A**, 224 (1968).

[6] W. E. Masker and R. D. Parks (to be published). We wish to thank Professor Parks for informing us of his results prior to publication.

find[7]

$$\sigma' \equiv \sigma - \sigma_N = (e^2/16\hbar d\tau) G(T) \qquad (1)$$

and

$$G(T) = \sum_{n=0}^{\infty} \frac{1}{1+n^2(\pi\xi/d)^2} = \frac{1}{2}\left[1 + \frac{d}{\xi}\coth\frac{d}{\xi}\right], \qquad (2)$$

where σ is the conductivity, σ_N is its normal-state value, $\tau \equiv (T-T_c)/T_c \ll 1$, and $\xi = 0.85(\xi_0 l/\tau)^{1/2}$ is the temperature-dependent coherence length in the dirty limit $\xi_0 \gg l$ [ξ_0 is the BCS coherence length ($\sim$830 Å in Pb) and l is the mean free path].

Denoting the "sheet resistance" by $R^s = Rw/l$, where R is the measured resistance of a film of width w and length l, we obtain for the fluctuation rounding of the resistive transition[8]

$$\frac{R_N - R(T)}{R(T)} = \frac{\Delta R}{R} = \frac{e^2 R_N{}^s}{16\hbar\tau} G(T) \equiv \frac{\tau_0{}^{\mathrm{AL}}}{\tau} G(T), \qquad (3)$$

for $\Delta R/R \ll 1$.

In the experimental data published previously and those presented here, the approximations used to obtain Eq. (3) are not always met. It is therefore important to understand under what conditions Eq. (3) is supposed to hold.

A. d/ξ Corrections to the Thin-Film Limit

Figure 1 shows the variation of G with d/ξ. In the limit of a "two-dimensional film" (i.e., $d/\xi \ll 1$), $G \to 1$. This is the limit which is normally assumed in the analysis of the data. As $\Delta R/R \to 0$ (i.e., at high temperatures), ξ decreases, and G becomes a temperature-dependent quantity greater than unity. At $d/\xi = 1$ the corrections to the two-dimensional theory are significant ($G = 1.17$), and for $d/\xi > 2$ three-dimensional behavior occurs to high accuracy.[7] In the three-dimensional limit ($d/\xi \gg 1$), we obtain

$$\frac{\Delta R}{R} = \frac{\tau_0{}^{\mathrm{AL}}}{2\tau}\left(\frac{d}{\xi}+1\right) \propto \tau^{-1/2}.$$

We shall note later the failure to see this behavior when expected in all reported data.

The breakdown of the thin-film approximation ($d \ll \xi$) may not be evident in the analysis of the data, however. The fractional difference in R/R_N between the exact and thin-film formulas is given by

$$\frac{\Delta(R/R_N)}{R/R_N} = \frac{R_N - R}{R_N}\left(1 - \frac{1}{G}\right). \qquad (4)$$

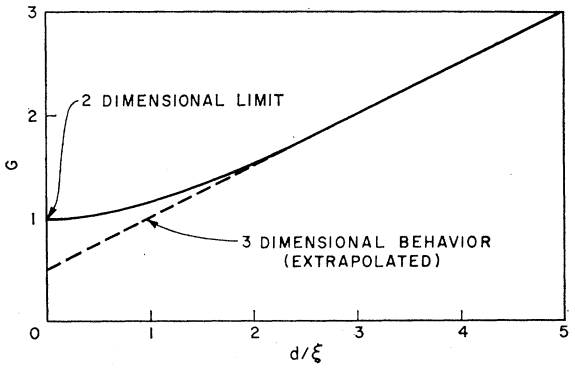

FIG. 1. G versus d/ξ showing film thickness corrections to the width factor [Eq. (2)].

Therefore the fractional error in the experimental determination of R/R_N must be less than that given by Eq. (4), if the finite thickness corrections are to be seen. When these corrections are small ($\tau \gg \tau_0$, $d \lesssim \xi$), the right-hand side of Eq. (4) will be substantially temperature-independent, and an erroneous value of R_N may mask the expected finite thickness corrections. We have found that a good least-squares fit using the function $\Delta R/R = \tau_0'/\tau'$ [$\tau' = (T-T_c')/T_c'$] can often be made of Eq. (3) for values of G up to 3 by small changes in the fitted values of τ_0 and T_c. In this fitted equation, however, the width factor τ_0' must be larger than $\tau_0{}^{\mathrm{AL}}$.

B. τ_0/τ Corrections and the Definition of T_c

The value of T_c in the preceding theoretical formulas is the transition temperature in the absence of fluctuations and is not necessarily that temperature at which the observed resistance vanishes. Furthermore, the expressions for σ' and $\Delta R/R$ in Eqs. (1) and (3) are only the leading terms in an expansion in $\tau_0/\tau \ll 1$. As σ' (or $\Delta R/R$) increases, higher-order terms in τ_0/τ become important. These have the effect of "renormalizing" T_c, and of changing the temperature dependence of R. In order to estimate the effect of the correction terms, let us represent $\Delta R/R$ in the form

$$\frac{\Delta R}{R} = \frac{\tau_0}{\tau}\left(1 + A\frac{\tau_0}{\tau}\right), \qquad (5)$$

where A is a constant, and in τ we use the "observed" T_c.[9] (We have set $G = 1$ for simplicity.) In fact, Eq. (5) is a rather accurate representation of the data at all

[7] See also H. Schmidt, Z. Physik **216**, 336 (1968), Eq. (28). Schmidt's statement that on the basis of his Eq. (28) [our Eq. (2)] the behavior is two-dimensional for $d/\xi \lesssim 3$ is incorrect (see Fig. 3 and Sec. II C).

[8] Equation (3) is sometimes replaced with $\Delta R/R_N = \tau_0/\tau$, anticipating, for the moment, that $G \approx 1$ in many experiments. By setting $T_c = T_c'(1+\tau_0{}^{\mathrm{AL}})^{-1}$ in the latter equation we obtain $\Delta R/R = \tau_0(1+\tau_0{}^{\mathrm{AL}})^{-1}/\tau'$, which is equivalent to Eq. (3) except for slight ($\tau_0 \sim 10^{-3}$) shifts in T_c and τ_0.

[9] By the "observed" T_c we mean the one at which the resistance vanishes in the exact theory. Since the experimental data are complicated by erratic behavior of the resistance very near T_c, we shall use for the observed T_c that value obtained by extrapolating the reproducible behavior at $R/R_N > 0.2$ down to $R/R_N = 0$.

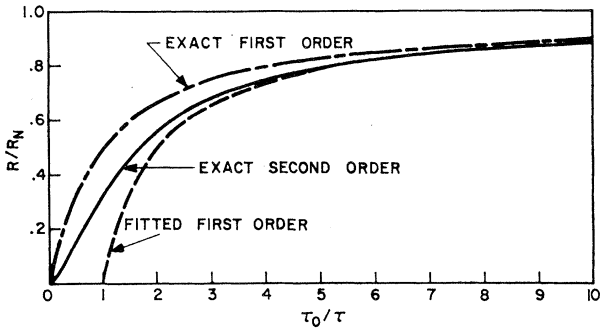

FIG. 2. R/R_N versus τ_0/τ, from Eq. (5), showing the effect of the second-order term. The "exact first-order" curve has $A=0$; the "exact second-order" curve has $A=1$. The "fitted first-order" curve represents Eq. (7) with $A=1$.

temperatures, except very near T_c, where the experiments are complicated by erratic behavior. Equation (5) may be rewritten so as to eliminate the second-order term in the following manner:

$$\frac{\Delta R}{R}=\frac{\tau_0}{\tau}\left(1+A\frac{\Delta R}{R}\right)+O\left[\left(\frac{\tau_0}{\tau}\right)^3\right], \tag{6}$$

$$\frac{\Delta R}{R}\simeq\frac{\tau_0(1+A\tau_0)^{-1}}{\tau'}, \tag{7}$$

where $\tau'=(T-T_c{}')/T_c{}'$ and $T_c{}'=T_c(1+A\tau_0)$. In Eq. (7) we have neglected terms of order $(\tau_0/\tau)^3$. Thus, by a slight adjustment of T_c the second-order term in Eq. (5) can be eliminated (to order τ_0/τ), the width factor remaining essentially unchanged (τ_0 is typically of order 10^{-3}). These results are illustrated graphically in Fig. 2, where Eq. (5) for R/R_N is given by the solid line, with the choice $A=1$. The first-order behavior [$A=0$, in Eq. (5)] is a good fit only for $R/R_N>0.9$. However, by shifting T_c by τ_0T_c we obtain the fitted first-order curve of Eq. (7) which is a good fit for $R/R_N>0.75$. A consequence of this behavior is that in fitting the data to the theory by adjusting T_c the fitted value of T_c will not be the observed T_c, but rather that which reduces the second-order corrections. The difference between the fitted and observed values of T_c, $T_c{}'-T_c=A\tau_0T_c$, gives the second-order coefficient. This coefficient may be estimated from the data by using the observed T_c, and the analysis is carried out in Sec. IV.

From Fig. 2 we see that a positive coefficient A, which gives $T_c{}'>T_c$, also introduces an inflection in the R/R_N-versus-T curves. This is the behavior generally observed experimentally. Any attempt to obtain a better fit for the smaller values of R/R_N by adjusting τ_0 (and T_c) will give values of τ_0 larger than the theoretical value $\tau_0{}^{\mathrm{AL}}$. For negative A, $T_c{}'<T_c$, and no inflection occurs.

C. Mean-Free-Path Corrections

For a clean film with a mean free path $l\gg d$, in the thin limit ($d\ll\xi$), the right sides of Eqs. (1) and (3) should be multiplied[3] by $(2l/d)[\ln(E_F/k_BT_cdk_F\tau)]^{-1}\approx l/6d$. We assume that the correction factor decreases monotonically to unity for $l\ll d$.

D. Analysis with No Fitted Parameters

The approximations $\xi\gg d$ and $l\ll d$ can fail although the data may appear reasonably fitted to a τ^{-1} dependence using a $\tau_0>\tau_0{}^{\mathrm{AL}}$. Since the T_c in Eqs. (1) and (3) is generally not the temperature at which the resistance is observed to vanish, its value cannot be obtained independently in the experiment. However, an analysis of R versus T can be made independently of T_c using the derivative dR/dT. In order to test the exponent of τ, let us represent $\Delta R/R$ in the form

$$\frac{\Delta R}{R}=\left(\frac{\tau_0}{\tau}\right)^n, \tag{8}$$

and differentiate, obtaining

$$\frac{1}{R^2T_c}\frac{dR}{d\tau}=\frac{1}{R^2}\frac{dR}{dT}=\frac{n}{\tau_0T_cR_N}\left(\frac{R_N-R}{R}\right)^{1+1/n}. \tag{9}$$

A log-log plot of $R^{-2}(dR/dT)$ versus $(R_N-R)/R$ gives a straight line of slope $1+1/n$ and an intercept of $(\tau_0T_cR_N/n)^{-1}$. For one-, two-, and three-dimensional systems, $n=\frac{3}{2}$, 1, and $\frac{1}{2}$, respectively. If R_N is determined independently, this result involves no fitted parameters. This plot will also show if a transition from two- to three-dimensional behavior occurs over the range of temperatures where the data are analyzed. An illustration of such a transition is shown in the upper curve of Fig. 3, where we plot Eq. (3) in derivative form for a hypothetical case where such a transition occurs near $\Delta R/R=0.03$.

III. EXPERIMENTAL

A. Film Preparation

Films were obtained by argon getter sputtering[10] of high-purity Pb at 4–6 Å/sec onto glass substrates held at ~80°K. The glass substrates were used to avoid the 500–1000 Å grooves normally found on polished single-crystal substrates. The sputtering rates were determined by weight gain measurements of thick (~5000-Å) films and by angstrometer measurements of thin (250-Å) films. These two rate determinations agreed to within 20%. For our studies, film thicknesses in the range 100–200 Å were used.[11] Films of less than 70 Å were not electrically continuous and films of more than 200 Å

[10] H. C. Theuerer and J. J. Hauser, Trans. Met. Soc. AIME **233**, 588 (1965).

[11] The film thickness, based on sputtering times, is uncertain for several reasons. It is not known how accurately the mean sputtering rates apply for the first 100 Å. Any oxidation which occurs will reduce the metallic film thickness from the values quoted above. It would appear likely for both reasons that the quoted film thicknesses are upper limits.

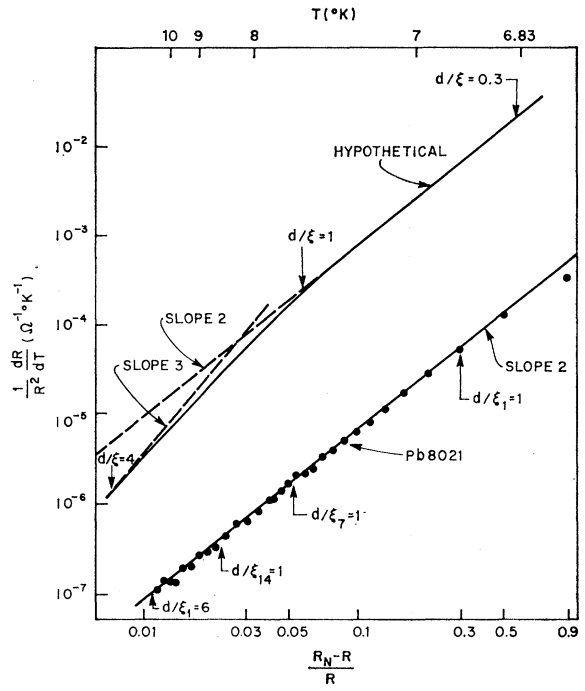

FIG. 3. $\ln(R^{-2}dR/dT)$ versus $\ln[(R_N-R)/R]$. In the upper curve the theoretical expression given in Eq. (3) is plotted for a hypothetical case where the transition from two- to three-dimensional behavior occurs in the vicinity of $(R_N-R)/R=0.03$. In the lower curve the data are plotted for sample 8021 (experimental errors are of the order of the scatter in the data). Several values of d/ξ are indicated by the arrows: ξ_1, ξ_7, and ξ_{14} are calculated assuming $l=0.3$, 7×0.3, and 14×0.3 Å, respectively. Slopes 2 and 3 are expected theoretically for the two- and three-dimension regimes.

had resistivities and τ_0 values too small for accurate measurement.

After deposition, the substrates, uniformly covered with the Pb film, were warmed to room temperature. Sample shapes suitable for standard four-terminal resistance measurements were obtained by scribing away a thin line of the Pb film on the substrate face. This was done to avoid the penumbra edge effects which can occur with the use of sample masks during deposition. Sample dimensions were usually 5.5 (between potential leads) $\times0.8$ mm.

B. Electron and Optical Microscopy of the Films

A crucial assumption in the application of Eq. (3) is that the current density is uniform throughout the sample (i.e., that $R_N^s=R_Nw/l$). We have investigated the uniformity of our films by electron microscopy.[12] The electron transmission studies[13] show that 90–95% of the film is continuous down to a scale of ~50 Å. No thickness variations were found, although transmission mission studies are not capable of detecting small variations of thickness. The uniformity of the average transmission intensity sets an upper limit of thickness variations of less than a factor of 2, on a scale of 100 Å and over a range of several microns (the largest distance traversed). The average grain size was ~30–40 Å. Electron diffraction showed the film to be mostly lead with some oxide and possible sulfide present. The remaining 5–10% of the film area is made up of randomly positioned small areas (~10–50 μ in size) within which the lead had spheroidized and no electrical continuity was possible. Since none of these areas was found to be less than ~10 μ, we were able to use the larger viewing field of an optical microscope (at 200–500×) and still see the 10–50-μ areas. No other irregularities on a scale of 1 μ or larger were seen and we conclude that the films were continuous over 90–95% of their areas. Further evidence of the uniformity of these films will be given in Sec. V.

Since the grain size in the films is only about six unit-cell lengths (and possibly less for the thinnest high-resistance films), and the grain boundary is at least several unit cells in thickness, a large fraction of the volume is disordered material. Thus on a scale of several unit-cell lengths these materials are quasiamorphous with short mean free paths.[14] Nevertheless, the disordered material is still superconducting, with a T_c only slightly less than the bulk.[15] On the scale of the coherence length ξ, which is usually greater than several hundred angstroms, the material is uniformly disordered and the theory of AL should apply. The fact that the T_c of Pb is largely independent of grain size and film thickness makes it likely that the rounding of the resistive transition will be less influenced by variations in these parameters.

The wide range in film resistivities obtained in our experiments suggests that this property is critically dependent on small changes in the sample preparation. For several films the resistance was monitored during and after deposition. A decrease in the resistance occurred when the film was first warmed to room temperature, reaching as much as a factor of 20 for a thicker (150-Å) film. The resistance reduction is probably due to grain growth, which is more pronounced in the thicker films.

C. Film Stability

Several of the films showed an increase in resistance with time when left in air (but not in vacuum). It is possible that this aging is due to oxidation. For most films the resistance did not vary more than 10–20%

[12] We are fully indebted to P. A. Turner for performing the electron microscope studies.

[13] For the transmission studies, the film (~100 Å thick) was necessarily deposited on graphite. The difference in substrates allows some doubt in equating the film structures. However, the defects found in the graphite substrate films, as will be noted, are also those seen by optical microscopy on the glass substrate films.

[14] See J. J. Hauser and H. C. Theuerer, Rev. Mod. Phys. **36**, 80 (1964).

[15] Bi is a superconductor in the amorphous state with properties quite similar to those of bulk Pb.

over several days. Resistances at room temperature before and after a run generally agreed to 5% or better.

D. Electrical Measurements

Most resistance measurements were made using a current of 0.055 μA. Ohmic behavior was confirmed to better than 1% for currents up to five times this value (the maximum current used) at all temperatures except very close to T_c ($R/R_N<0.1$). Close to T_c a generally larger noise level and a diminishing signal level made it difficult to confirm Ohmic behavior with comparable certainty.

Voltages were measured to an absolute accuracy of 2–20 nV and relative accuracy of 10^{-3}–10^{-4}. For $R/R_N>0.5$ the measured resistances were generally reproducible on temperature cycling to within the estimated errors of the measurement. For $R/R_N<0.1$, however, the shape of the R-versus-T curves was often not reproducible even for temperature changes in the same direction. This behavior does not restrict the data analysis in accord with Eq. (3), since the theoretical result holds only for R/R_N approaching unity and the analysis was restricted to $R/R_N>0.5$.

For most measurements the sample was located in a large hydrogen-annealed mu-metal shield within which the measured magnetic field was less than 0.05 Oe. For one sample, data were also taken in parallel and perpendicular fields of 1000 Oe.

E. Temperature Control and Measurement

The temperature of the sample chamber (which contained a small amount of He exchange gas) was automatically regulated during measurement. The absolute accuracy of the temperature measurements, using a germanium resistance (control) thermometer, was estimated to be ± 10 mdeg. The relative measurement accuracy and the temperature stability provided by the controller were both ~ 0.1 mdeg. For the magnetic field measurements, the shift in the temperature scale for 10^3 Oe was ~ -1 mdeg.

IV. RESULTS

In analyzing the experimental data according to Eq. (3), the τ^{-1} dependence can be checked without multiple-parameter curve fitting by using Eq. (9), which requires dR/dT, R, and R_N and is independent of T_c. The observed values of R_N are obtainedby extrapolation of the small temperature dependence[16] (~ 0.1–0.5% per °K) at temperatures sufficiently above T_c for fluctuation effects to be negligible.[17] This determination

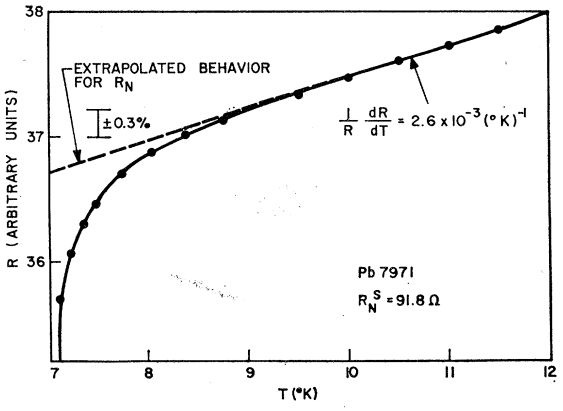

FIG. 4. R/R_N versus T for sample 7971 showing the temperature dependence at the higher temperatures. R_N was determined by the extrapolation of this behavior.

is shown for a typical case in Fig. 4. We show in the lower curve of Fig. 3 the plot of Eq. (9) for sample 8021. The slope of $1+1/n=2$ expected for fluctuations in a two-dimensional system is well confirmed for $0.5>\Delta R/R>0.01$. Thus it appears that the finite thickness corrections are negligible, and we take the thin limit $G=1$ in Eq. (3). In this film the d/ξ corrections should be very large. This important point is given a fuller discussion in Sec. IV B. From the intercept $\tau_0 T_c R_N$ of this plot we find $\tau_0=39.3\times10^{-3}=0.072\tau_0^{\mathrm{AL}}$ (using the observed T_c and $R_N=R_N^s l/w=38\,610\,\Omega$).

To obtain accurate values of T_c and τ_0 a least-squares fit of the data was made to the equation $\Delta R/R=\tau_0/\tau$, in which τ_0, R_N, and T_c are fitted parameters. One test of the validity of this analysis lies in comparing the fitted and observed values of R_N. In all cases these values agree to within the error (typically $\sim\pm0.3\%$) in determining R_N. The small temperature dependence of R_N is included in the analysis but it is of minor importance in determining the fitted values.

Figures 5–7 show R/R_N versus T for films with $R_N^s=5290$, 268, 5.00 Ω. The solid lines give R/R_N

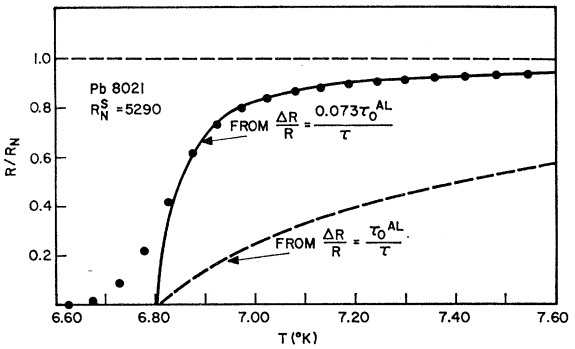

FIG. 5. R/R_N versus T for sample 8021. The solid line is from $\Delta R/R=\tau_0/\tau$, with fitted τ_0, T_c, and R_N. The dashed line is for the theoretical width factor τ_0^{AL}, using the same R_N^s and T_c.

[16] The films generally showed resistance ratios $\rho(300°\mathrm{K})/\rho(10°\mathrm{K})\sim 2$–4. For about half of the samples the mean free paths are so small that the temperature dependence would not arise from ordinary phonon scattering. This behavior is also seen in other films of thickness up to 10^5 Å. See Ref. 11 and J. J. Hauser and H. C. Theuerer, Phys. Rev. **134**, A198 (1964).

[17] In one or two cases R_N was also obtained by applying magnetic fields to remove the fluctuation effects. These values were in agreement with those obtained by the high-temperature extrapolation method.

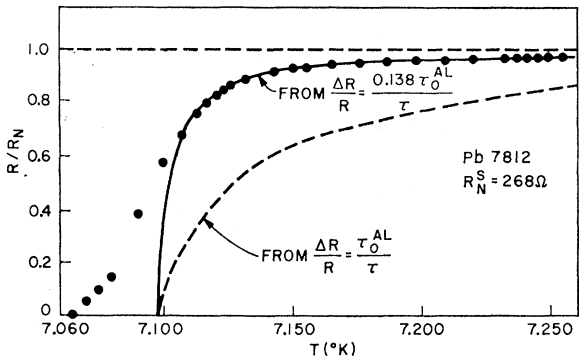

FIG. 6. R/R_N versus T for sample 7812. The solid line is from $\Delta R/R=\tau_0/\tau$, with fitted τ_0, T_c, and R_N. The dashed line is for the theoretical width factor τ_0^{AL}, using the same $R_N{}^s$ and T_c.

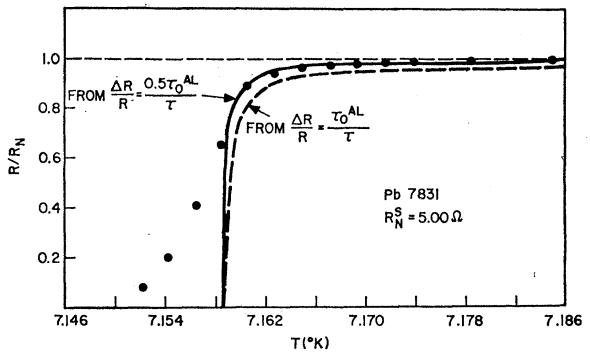

FIG. 7. R/R_N versus T for sample 7831. The solid line is from $\Delta R/R=\tau_0/\tau$, with fitted τ_0, T_c, and R_N. The dashed line is for the theoretical width factor τ_0^{AL}, using the same $R_N{}^s$ and T_c.

versus T, using the fitted τ_0, R_N, and T_c. Since the R_N and τ^{-1} determinations are made independently, only τ_0 and T_c are free variables. The dashed lines give the result for R/R_N versus T, using τ_0^{AL} and the same R_N and T_c. For the low-resistance film it is evident (see Fig. 7) that the fit must be made as close to $R/R_N=1$ as the experimental accuracy (in determining R_N) will allow. The fit shown in Fig. 7 could be improved around and below the knee of the curve by using a larger τ_0 and by shifting $T_c{}^f$. However, the resulting error in the high-temperature tail (where the theory applies), which may not appear large in Fig. 7, is well beyond the experimental error. The experimental width factors τ_0 are smaller than the theoretical value τ_0^{AL} by factors of 2–14. This discrepancy is clearly outside of all experimental errors.

In all cases the fitted $T_c{}^f$'s were higher than the temperatures at which the resistance vanished. We can estimate the value of the coefficient A used in Eq. (7) from the relation

$$A=(T_c{}^f-T_c)/\tau_0 T_c.$$

The error in A is about a factor of 2 (because of the error in the determination of T_c [9]), with the largest uncertainties occurring for films with small $R_N{}^s$. For most films $A\sim 5$–10, which is reasonably constant considering the fact that $R_N{}^s$ varies by three orders of magnitude. The large value of A also shows that the $\Delta R/R$ analysis must be restricted to $R/R_N>0.7$. Indeed, for the low-resistance films (e.g., Fig. 7), if the least-squares fit is restricted to $0.5<R/R_N<0.9$, one obtains $\tau_0>\tau_0^{AL}$.

In Table I we give the results for a number of samples with $R_n{}^s$ between 2.5 and 5300 Ω. The mean free paths have been calculated from $\rho^{-1}=Ne^2l/mv_F$, where $N=2.54\times 10^{22}$ (electrons and holes)/cc, and $\langle mv_F\rangle$

TABLE I. Properties and data for lead films.

Sample No.	Nominal thickness (Å)	$R_N{}^s$ (7.5°K) (Ω)	l (7.5°K) (Å)	$T_c{}^f$ [a] (°K)	τ_0/τ_0^{AL}	ρ(film)/ρ(bulk) (at 300°K)
8041	150	2.57	390	7.186	0.48 ±0.1	1.5
7831	125	5.00	240	7.158	0.56 ±0.1	1.10
7771	200	6.50	115	7.200	0.50 ±0.15	4.5
7851	125	7.18	165	7.177	0.60 ±0.1	1.65
7971	100	91.8	16	6.994	0.35 ±0.1	15
7881	100	125	12	7.004	0.62 ±0.05	17.5
7972[b]	100	246	6	6.930	0.33 ±0.05	34.5
7812	100	268	5.5	7.099	0.138±0.03	57
7921[c]	?	678	···	6.624	0.50 ±0.1	···
8021	100(?)	5290	0.3	6.809	0.073±0.005	465

Data for sample 7881 in 10^3 Oe

	$T_c{}^f$ (°K)	τ_0/τ^{AL}
$H\parallel$film plane	6.990	0.76±0.1
$H\perp$film plane	6.876	0.89±0.1

[a] Typical uncertainties in $T_c{}^f$ are $<(2\times 10^{-3})$°K for $R_N{}^s<7.2$ Ω and $<(5\times 10^{-3})$°K for $R_N{}^s\geq 92$ Ω.
[b] Sample 7972 showed the aging effect discussed in Sec. III. The original sample (7971) increased in resistance by a factor of ~2.7 when left in air at 300°K for about 7 h.
[c] Sample 7921 was inadvertently sputtered from a dirty target. The sputtering time was longer and the T_c lower than for any other sample. Its data are questionable and are included for completeness only.

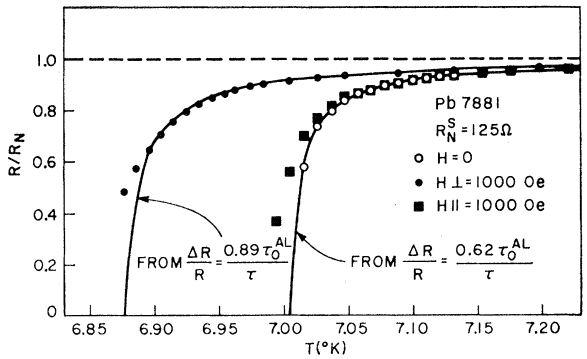

FIG. 8. R/R_N versus T for sample 7881 in a magnetic field. The solid lines are the fitted curves for zero field and H perpendicular to the film plane. No fitted curve is shown for the parallel field data.

$\approx 10^{-19}$ g cm/sec.[18] This gives

$$l = (1.54\times 10^{3})\rho^{-1}, \tag{10}$$

with l in Å and ρ in $\mu\Omega$ cm, which is in good agreement with the experimental result obtained by Cody and Miller[19] for thin Pb films. The errors in the fitted values of τ_0 represent our estimate of the combined effect of the experimental errors in R, R_N, and T, and the sensitivity of the fit to the range in R/R_N over which the data were analyzed.

For sample-7881 measurements were made in a magnetic field. In Fig. 8 we give the results obtained in a field of 10^3 Oe applied perpendicular to the current, and both parallel and perpendicular to the film plane.[20] For H parallel to the film the R/R_N-versus-T curve is shifted to lower temperatures and slightly broadened. The data can still be represented reasonably well by the equation $\Delta R/R = \tau_0/\tau$ and the fitted values of τ_0 and T_c are given in Table I. The changes in these values are within their respective uncertainties. For H perpendicular to the film plane the changes were larger:

$$\frac{dT_c}{dH_\perp} = -1.3\times 10^{-4}\ ^\circ\mathrm{K/Oe} \quad \text{and} \quad \frac{1}{\tau_0}\frac{d\tau_0}{dH_\perp} \approx 2\times 10^{-4}\ \mathrm{Oe}^{-1}.$$

Our experimental results can be summarized as follows: (a) In every case $\Delta R/R$ shows a τ^{-1} temperature dependence. (b) The fitted T_c's are always higher than the temperature at which the resistance vanishes. For $R/R_n < 0.7$ the observed values of R/R_n are usually greater than the fitted curves, and an inflection occurs at $R/R_N \sim 0.3$. This behavior can be represented by a positive second-order τ_0/τ term [see Eq. (5) and Fig. 2]. (c) The measured width factor τ_0 is less than the theoretical value τ_0^{AL}. (d) The measured width factor is not proportional to $R_N{}^s$, although τ_0 generally increases with increasing $R_N{}^s$. (e) For sample 8021, although $d/\xi > 1$ over most of the experimental range in R/R_N, the expected three-dimensional behavior ($\Delta R/R \propto \tau^{-1/2}$) was not observed.

V. DISCUSSION

The analysis of the fluctuation rounding of the resistive transition often assumes that the observation of the τ^{-1} dependence is a reliable indication of the intrinsic effect calculated by the theory. Since all reported data[1,4–6] confirm the τ^{-1} dependence but not the theoretical value of τ_0, the temperature dependence may not be a sufficient condition for quantitative comparison. In cases where the observed τ_0 is too small, of course, the discrepancy cannot easily be ascribed to extraneous mechanisms, since most of these are expected to add to the intrinsic widths. Effects which broaden the transition are numerous; however, there is little basis for predicting their temperature dependence in these thin dirty films. The data also fail to show the departures from two-dimensional behavior when predicted by theory. In view of these discrepancies it is necessary to review quite carefully the possible sources of error, either in the experiments, or in the theory, or, finally, in the application of theory to experiment.

A. Experimental Sources of Error

The most likely experimental source of error leading to small values of τ_0 is nonuniform current flow.[21] We have discussed the microscopic observations of the films indicating homogeneity and continuity over 90% of the area. Additional evidence lies in the data of sample 7831. For this sample the room-temperature resistivity Rwd/l was 24×10^{-6} Ω cm, compared to the value 22×10^{-6} Ω cm for bulk lead. The factor of 2 discrepancy between the experimental and theoretical values of τ_0 cannot be attributed to incorrect values of w/l, since this would imply that the 125-Å sputtered film had one-half the resistivity of the bulk. Similar arguments for sample 8041 also would lead to a resistivity less than that of bulk. Further evidence of uniform current flow will be presented in the discussion of the magnetic field effects.

There are several ways in which the observed width can be greater than the true width while the resistive rounding still appears to have a τ^{-1} dependence. For example, irregularities in film thickness may lead to erroneously small values of $R_N{}^s$ and large values of τ_0. In Sec. II it was shown that the failure of either of the approximations $l \ll d$ or $d \ll \xi$ leads to widths that are larger than that predicted by the thin-film theory, although a reasonable τ^{-1} dependence can often be obtained over a factor of 5 or more in τ by small adjustments in R_N and T_c.

[18] J. R. Anderson and A. V. Gold, Phys. Rev. **139**, A1459 (1965).
[19] G. D. Cody and R. E. Miller, Phys. Rev. **173**, 481 (1968).
[20] Measurements at higher fields are complicated because the shift in the germanium resistance temperature calibration ($\sim$1 mdeg at 10^3 Oe) goes as H^2.
[21] The error arises when the ratio w/l for the current flow is less than that of the film dimensions (e.g., in the presence of large nonconducting areas), which leads to a calculated $R_N{}^s$ greater than the true value.

An erroneously large value of τ_0 can also result from analyzing the data too close to T_c. As discussed earlier, by carrying out the analysis in the range $0.5<R/R_N<0.9$ for films of low R_N^s (see Fig. 7), we have found $\tau_0 \gtrsim \tau_0^{AL}$. This, we believe, explains why Smith *et al.*[5] find $\tau_0=\tau_0^{AL}$ in Pb films with $R_N^s \approx 3$–$15\ \Omega$, from analyses over this restricted range.

The temperature dependence of any additional width from extraneous sources (e.g., film irregularities) cannot be estimated. In the data of Strongin *et al.*[4] for Al films, a τ^{-1} dependence is observed for the film with $l=5$ Å, where the approximations $l \ll d \ll \xi$ are well satisfied, but the width factor $\tau_0 \approx 2.5\tau_0^{AL}$. Furthermore, Parks *et al.*[6] find $\tau_0 > \tau_0^{AL}$ over a wide range in R_N^s for Al films. If the theory is correct, there must exist additional broadening mechanisms which show the same τ^{-1} dependence. If this be true, it would be extremely difficult to establish experimentally that the measured width is that minimum value due to intrinsic fluctuations, which the theory calculates. However, we believe that our results, which show widths *less* than the theoretical value, indicate that the theory is not correct for the determination of τ_0.

B. Failure to See Three-Dimensional Behavior

In the Al data of Strongin *et al.*[4] for the sample with $l=0.2$ Å, $d/\xi=1$ at $R/R_N \approx 0.3$ and $d/\xi=2$ at $R/R_N=0.83$, so that throughout most of the experimental range the behavior should be three-dimensional. For Glover's[1] sample, the corrections are less important at comparable $\Delta R/R$. Assuming ξ_0 in Bi to be the same as in Pb[15], and $l=5$ Å, then $\xi=d$ ($d=470$ Å) when $R/R_N=0.95$. Since G increases rapidly as τ increases beyond its value (10^{-2}) at this point, the agreement between the thin-film theory ($G=1$) and the data out to $\tau=1$ is surprising.[22] The excellent fit to a τ^{-1} dependence at $\tau \sim 0.3$, for example, would imply $d/\xi \lesssim 1$ at that temperature, from which one calculates $\xi_0 l \gtrsim 10^5$ Å^2. This is larger than our estimate for this parameter by about one order of magnitude. However, assuming the more reasonable estimate $\xi_0 l = 10^4$ Å^2, then, from Eq. (4), it is seen that a fractional error (underestimate) in the determination of R_N by 0.16% would change the observed τ^{-1} dependence to the predicted behavior [Eq. (1)].

Finally, the failure to see three-dimensional behavior in our Pb films is clearly demonstrated by the data of sample 8021. In the derivative plot of this data (Fig. 3, lower curve) we note that $d/\xi \sim 1$ at $\Delta R/R=0.3$. Thus throughout most of the range where the analysis is made we should expect three-dimensional behavior. However, somewhat similar to Glover's results, we find an excellent fit to the thin-film formula ($\Delta R/R \propto \tau^{-1}$) out as far as $\tau \sim \frac{1}{2}$. Since our estimate of ξ is subject to error, we also show in Fig. 3, as indicated by ξ_7 and ξ_{14}, the points at which $d/\xi=1$, assuming mean free paths *larger* by factors of 7 and 14, respectively, than those calculated from R_N^s. Thus, if the small observed width factor $\tau_0^{AL}/14$ for this film is to be explained by assuming some effective l which is larger (or R_N^s smaller) by a factor of 7 or 14, then three-dimensional behavior would still be expected for part of the data. Hence, a consistent and puzzling discrepancy between experiment and theory for Bi, Al, and Pb is the failure to see three-dimensional behavior when predicted. Because of the large discrepancy in at least one case, this conclusion remains true even if our particular interpolation formula [Eq. (2)] or our estimates of ξ_0, l, and d are not accurate.

C. Magnetic Field Effects

In a thin film, the perpendicular critical field is H_{c2}, given by the relation

$$H_{c2}=H_\perp=\sqrt{2}\kappa H_c(T), \tag{11}$$

where κ is the Ginzburg-Landau parameter and $H_c(T)$ is the thermodynamic critical field. In the dirty limit, we may use the Ginzburg-Landau-Abrikosov-Gor'kov (GLAG) theory[23] to express Eq. (11) in terms of the mean free path and measured quantities[24]:

$$H_\perp=0.87\frac{\lambda_L(0)}{l}H_c(T)=\frac{0.22\varphi_0\tau}{\xi_0 l}, \tag{12}$$

where $\lambda_L(0)$ is the London penetration depth and $\varphi_0=hc/2e$ is the flux quantum. Since in our films T_c is close to its bulk value, we assume that other thermodynamic quantities such as ξ_0 H_c and $\lambda_L(0)$ are also equal to their experimental values for bulk Pb. Using $\xi_0=830$ Å $\lambda_L(0)=370$ Å and $(dH_c/dT)_{T=T_c}=-205$ Oe/°K, we obtain

$$-\left(\frac{dH_\perp}{dT}\right)_{T=T_c}=-\sqrt{2}\kappa\left(\frac{dH_c}{dT}\right)_{T=T_c}=\frac{7\times10^4}{l}\ \text{Oe/°K}\quad\text{(calculated)} \tag{13}$$

(with l in Å), to within ±5% using either formula in Eq. (12). Cody and Miller[19] have obtained experimentally for Pb films (for $d>400$ Å) $\kappa=235/l$ (l in Å), which implies

$$-\left(\frac{dH_\perp}{dT}\right)_{T=T_c}=\frac{6.8\times10^4}{l}\ \text{Oe/°K}\quad\text{(experimental)}, \tag{14}$$

[22] This has been pointed out by E. Abrahams and J. W. F. Woo [Phys. Letters **27A**, 117 (1968)], who also suggest that for large τ we should put $\tau \to \ln(T/T_c)$.

[23] C. Caroli, P.-G. de Gennes, and J. Matricon, Physik Kondensierten Materie **1**, 176 (1963).

[24] P.-G. de Gennes, Physik Kondensierten Materie **3**, 79 (1964).

in good agreement with Eq. (13) calculated from the GLAG theory.[25]

For sample 7881, $(dH_\perp/dT)_{T=T_c}=-6\times10^3$ Oe/°K (where we have used the observed T_c rather than the fitted value). By Eq. (14) this gives $l=11$ Å, in agreement with the value $l=12$ Å obtained from the measured $R_N{}^s$. This agreement provides further evidence that the measured $R_N{}^s$ was not in error due to nonuniform current flow.

D. Sources of Error in the Theory

The main results of AL which are not borne out by the experimental data are the independence of σ' on mean free path in the thin-film limit, and the transition from two- to three-dimensional behavior. We shall now examine the physical basis for these results of the theory, in an effort to elucidate the possible causes for their breakdown.

1. Independence of σ' on Mean Free Path

The general expression for the conductivity [Eq. (14) of Ref. 3] involves the vertex C, the constant η related to the coherence length ($\xi^2\equiv\eta\tau^{-1}$), and the relaxation rate of the order parameter $\Omega_\psi\equiv(8k_B/\pi\hbar)(T-T_c)$ [see Eq. (11) of Ref. 3, and Refs. 7 and 22]. Two of these quantities (C and η) are related to static properties and enter into the time-independent GLAG theory, while the third (Ω_ψ) refers specifically to the time dependence of the order parameter. For thin dirty films ($l\ll d\ll\xi$), AL[3] assume that σ' is obtained by simply restricting the integral in their Eq. (14) to the coordinates in the plane of the film, with C, η, and Ω_ψ having the same values as for dirty bulk samples. The conductivity σ' is then proportional to $(C^2/\eta^2)\Omega_\psi{}^{-1}$ [see Eq. (17) of Ref. 3], which is independent of the mean free path in the dirty limit.

That C/η does not depend on l may be verified from the expressions quoted in Eqs. (6) and (12) of AL, but it follows also from the more general version of the GLAG theory given by Caroli *et al.*,[23] which is valid for very dirty materials in which $(k_Fl)^{-1}$ need not be small compared to unity.[26] Moreover, since C and η are related[23,26] to the penetration depth λ and upper critical field H_{c2}, it is in principle possible to verify experimentally that C/η is independent of l.

The assertion that Ω_ψ depends only on temperature, and not, for instance, on l, has to our knowledge never been verified experimentally, and may not be true in all cases. To the extent that the equilibrium GLAG theory is assumed to hold, the present results may be taken as an indication that Ω_ψ does depend on the parameters of the system and not just on $T-T_c$. On the other hand, it does not seem likely that all the data can be explained simply by a variation of relaxation rate from sample to sample, since, for comparable values of k_Fl and d/ξ, the AL theory overestimates the fluctuations in Pb, underestimates them in Al,[4,6] and yields the correct answer in Bi.[1]

2. Change from Two- to Three-Dimensional Behavior

The failure to observe three-dimensional behavior experimentally when expected ($d/\xi\gtrsim1$) suggests that if the AL theory is correct, the coherence length may be larger than that given by the GLAG expression $\xi=0.85(\xi_0l/\tau)^{1/2}$. It is possible that not all processes which limit the mean free path in the normal state act to restrict the coherence length in the superconducting state for very dirty films. Physically this might lead to smaller fluctuation effects than those calculated using the mean free path obtained from the film resistance, and thus explain reduced values of τ_0.[27] In any case, it follows from Eq. (12) that the perpendicular critical field near T_c is given by

$$H_\perp=\varphi_0/(2\pi\xi^2)\,, \tag{15}$$

so that the coherence length ξ for $T<T_c$ can be determined by an independent measurement involving equilibrium properties, and directly compared to d. The observation of a *difference* in temperature dependence of σ' in going from $d<\xi$ to $d>\xi$ would constitute an experimental determination of ξ for $T>T_c$. This important observation would, in our opinion, be the best proof that intrinsic fluctuations dominate the measured resistive transition. It would also represent the first experimental evidence that the Ginzburg-Landau theory may be used to describe not only the average order parameter, but also fluctuations from this average. The most promising materials in which to observe such a change in behavior are amorphous Bi and granular Al, since relatively thick films may be made with short mean free paths, so that the condition $d=\xi$ occurs where fluctuations are still measurable. The measurement of $H_\perp$ on the same sample will, via Eq. (15), also test the symmetry of the coherence length about T_c, which is assumed in the Ginzburg-Landau theory.

3. Strong Coupling Effects

Aslamazov and Larkin[3] used the weak coupling BCS theory of superconductivlty, and it is natural to ask what modifications will arise due to the strong coupling

[25] For strong coupling materials, Eq. (12) may contain some modifications [see G. Eilenberger and V. Ambegaokar, Phys. Rev. **158**, 332 (1967)]. We assume that these are adequately taken into account by using *experimental* values for H_c, ξ_0, and $\lambda_L(0)$ rather than the BCS values. Since the κ values observed in Ref. 19 agree with Eq. (13), this assumption appears to be correct.

[26] Indeed, the coefficient C of AL is the same as T of Ref. 23 [Eq. (3.11)], and η of AL corresponds to $L[gN(0)]^{-1}$ [Eq. (3.9)]. As shown in Ref. 23 by general sum-rule arguments, both L and T are proportional to the normal-state conductivity σ_N, so that their ratio is independent of the mean free path.

[27] Although in two dimensions the AL theory predicts that σ' is independent of l (and ξ), it is difficult to estimate from the theory what effect a breakdown of the relation $\xi=0.85(\xi_0l/\tau)^{1/2}$ would have on σ'. It seems physically reasonable to us that a larger ξ will lead to smaller fluctuation effects.

nature of the materials used in many experiments (Pb and Bi). The extension of the AL theory to strongly coupled materials has been reported by Fulde and Maki,[28] who find that τ_0 should be divided by a factor α, related to the slope of H_{c2} at T_c, and estimated to be 2 in Pb. The strong coupling theory, however, which still shows τ_0 proportional to R_N^s (as long as T_c does not vary), is not in agreement with our findings in Table I. It is possible that for the low-resistance films ($R_N^s < 100\ \Omega$) the observed discrepancies are just due to strong coupling effects, and therefore the theory fails only for the high-resistance films. Again, however, existing data are in conflict. Amorphous Bi, which is also a strong coupling material,[29] does not show a reduced τ_0.

4. *Thermal Effects Associated with the Lattice*

Another source of discrepancy may lie in other lattice effects, which influence the temperature fluctuations in the system. In a recent paper Kadanoff and Laramore[30] used the mode-mode coupling theory including lattice effects to estimate the rounding of the resistive transition. When electrons and lattice are thermally uncoupled, these authors reproduced the answers of AL apart from numerical factors, which the mode-mode theory does not specify. Even in this uncoupled case, however, the mechanism for producing rounding is a combination of thermal and order-parameter fluctuations, which may not be identical to the AL mechanism, involving the propagation of fluctuating pairs.

When electrons and lattice are thermally coupled, the temperature fluctuations are controlled by the *total* specific heat (electronic and lattice), and the mechanism of Kadanoff and Laramore leads to reduced τ_0 values since the lattice can now damp the fluctuations. The formula obtained by these authors in the "mean-field" region ($\Delta R/R \ll 1$) can be written

$$\frac{\Delta R}{R} \approx \frac{\tau_0^{\mathrm{AL}}}{\tau} \frac{2}{2+L}, \tag{16}$$

where $L=0$ in the uncoupled case and

$$L=(r+1)C_L/C_e$$

in the coupled case. C_L and C_e are the lattice and electronic heat capacities, respectively, and $r=\kappa_L C_e/\kappa_e C_L$ is the ratio of lattice and electronic thermal diffusivities.[31] We estimate very roughly $r\sim 1$ at 7.5°K for the low-resistance films, and since $C_L/C_e \sim 25$ at this temperature for Pb, Eq. (16) would lead to a significant reduction in fluctuation effects. To estimate the degree of coupling Kadanoff and Laramore[30] compare the electronic fluctuation rate with some electron-phonon relaxation rate. The former is $\hbar^{-1}k_B T_c \tau \approx \tau \times 10^{11}$ sec^{-1}, which seems less than most electron-phonon relaxation rates for $\tau \lesssim 10^{-1}$, indicating large coupling effects. Thus the modification of the AL theory introduced in Ref. 30 might explain the narrow widths observed in our experiments.

In practice, however, this explanation seems to us unconvincing, for the following reasons: (a) The observed reduction factor is less than the specific heat ratio, and therefore we must assume some intermediate state of coupling, for which the observed τ^{-1} dependence is not obviously valid. (b) Amorphous Bi and Pb are very similar in their superconducting and phonon properties, so that Glover's[1] experiments should also show a reduced τ_0. (c) The lattice effect always leads to a *reduction* of the width and would not explain the *increased* widths observed in thick Al films.[6] (d) Finally, from a theoretical point of view, it seems to us that the rounding of the resistive transition should not depend so sensitively on the magnitude of temperature fluctuations. This is seen most clearly from the phenomenological formulation of the AL theory presented by Schmidt[7] and by Abrahams and Woo.[22] The width τ_0 is proportional to $\langle\Psi^2\rangle$, and it is $\langle\Psi^2\rangle$ which determines the jump in the specific heat at T_c. This jump, however, is not dependent upon C_p itself. The lattice only makes small corrections to the jump in specific heat, and to the order-parameter relaxation rate (which also enters the formulas of Schmidt,[7] and of Abrahams and Woo[22]), by renormalizing the electronic parameters. This is presumably the effect calculated by Fulde and Maki,[28] and only leads to a factor of 2 reduction, as mentioned earlier. On the other hand, it is important to note that these theoretical arguments are only suggestive, and it would be desirable to study by a microscopic calculation which includes the lattice,[32] the effect of temperature fluctuations and energy transport on resistive fluctuations.

5. *Fluctuation Effects in a Magnetic Field*

In the presence of a magnetic field, the effect of fluctuations on the resistance was estimated by AL by simply taking into account the dependence of T_c on H. Schmidt[33] has recently proposed that additional fluctua-

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[28] P. Fulde and K. Maki, in *Proceedings of the Eleventh International Conference on Low-Temperature Physics*, edited by J. F. Allen, D. M. Finlyson, and D. M. McCall (University of St. Andrews Printing Department, St. Andrews, Scotland, 1969), Vol. I, p. 120; and (to be published).

[29] J. T. Chen, T. T. Chen, J. D. Leslie, and H. J. T. Smith, Phys. Letters **25A**, 679 (1967).

[30] L. P. Kadanoff and G. Laramore, Phys. Rev. **175**, 579 (1968). We wish to thank these authors for sending us a copy of their paper prior to publication, and for discussing their theory with us.

[31] We have assumed that the electrical and thermal diffusivities of the electron gas are equal. This is expected for free-electron behavior.

[32] For $T<T_c$ the time dependence of the order parameter was considered microscopically in the presence of phonon effects by E. Abrahams and J. W. F. Woo, Phys. Rev. **169**, 407 (1968).

[33] H. Schmidt, Phys. Letters **27A**, 658 (1968); and (personal communication).

tions will arise in the presence of a depairing mechanism, such as paramagnetic impurities. In the thin-film limit Schmidt found

$$\frac{\Delta R}{R}=\frac{\tau_0^{\mathrm{AL}}}{\tau}f(\rho), \tag{17}$$

where[33,28]

$$f(\rho)=\psi'(\rho+\tfrac{1}{2})\{\psi'(\tfrac{1}{2})[1-\rho\psi'(\rho+\tfrac{1}{2})]\}^{-1}. \tag{18}$$

Here $\rho=\hbar(2\pi k_B T_c\tau_s)^{-1}$ is the depairing parameter and ψ' is the derivative of the digamma function. Now it is well known that a magnetic field is a depairing mechanism in thin films,[34] with parameters

$$\rho_{\|}=\frac{\pi}{36}\frac{\hbar v_F l}{k_B T_c\varphi_0^2}d^2H^2 \tag{19}$$

and

$$\rho_{\perp}=\frac{1}{6}\frac{\hbar v_F l}{k_B T_c}\frac{H}{\varphi_0} \tag{20}$$

for parallel and perpendicular fields, respectively. Thus we expect additional broadening in a magnetic field.[35] In order to estimate ρ we may use the relation[34]

$$\frac{\delta T_c}{T_{c0}}\equiv\frac{T_{c0}-T_c}{T_{c0}}=(\tfrac{1}{2}\pi^2)\rho+\cdots, \quad \text{for} \quad \rho\to 0. \tag{21}$$

The function $f(\rho)$ given in Eq. (18) has a small ρ expansion of the form

$$f(\rho)\simeq 1+1.5\rho, \tag{22}$$

which implies, using Eq. (21),

$$f(\rho)=1+0.3\frac{\delta T_c}{T_{c0}}=\frac{\tau_H}{\tau_0^{\mathrm{AL}}}, \tag{23}$$

where τ_H is the width in a magnetic field. Although our data are very limited, it appears from Table I that the observed broadening is larger than that predicted by Eq. (23).

For perpendicular fields the superconducting state of thin films[36] below $T_c(H)$ is analogous to the mixed state of type-II superconductors, with a regular array of quantized vortices.[37] In the latter case, additional fluctuation effects have been predicted[38] for $T<T_c(H)$ [or $H<H_{c2}(T)$], caused by the spatial degeneracy in the position of the vortices. It is possible that the same effect will reflect itself in fluctuations above $T_c(H)$, leading to an additional broadening of the resistive transition, over and above the one caused by the depairing effect of the magnetic field [Eq. (17)]. It is hoped that more detailed investigations, both experimental and theoretical, of the magnetic field dependence of the resistive transition will yield valuable information on the validity of the theory.

VI. SUMMARY AND CONCLUSIONS

Although we have briefly discussed several possible sources for the discrepancies between theory and experiment, there is no quantitative test of the correctness of any one of these. Even qualitatively, no explanation seems to be compatible with experimental data on all materials studied to date.

It is well to consider again the assumption of uniformity of current flow and sample geometry, because its proof is ultimately hindered by our ignorance of the detailed structure of our samples. We have presented evidence in several forms to show that our values of $R_N{}^s$ are correct for samples 7831, 8041, and 7881. For the other samples no comparable evidence is available. However, for the six films with $R_N{}^s$ between 2.5 and 125 Ω we find $\tau_0/\tau_0{}^{\mathrm{AL}}\sim 0.5\pm 0.15$ for five of them. It seems implausible that any error from nonuniform current flow should lead to such constant values of $\tau_0/\tau_0{}^{\mathrm{AL}}$ over such a wide range of $R_N{}^s$. Finally, from observations with an optical microscope we have found all films to be uniform on a scale of 1-μ. We have inferred that the scale of uniformity extends to 50 Å from observations with an electron microscope. We therefore conclude that, with the possible exception of sample 8021, the disagreement with the AL theory is not due to sample nonuniformiites.

In order to explain the discrepancies it is important to decide whether the AL theory is incorrect or whether its range of applicability must be limited by as yet unspecified criteria. Until this question has been clarified, the theory must be considered at best qualitatively correct in its prediction of a τ^{-1} temperature dependence for thin films and a relative transition width which increases with increasing $R_N{}^s$.

The major qualitative failure in the comparison of the AL theory to presently available data occurs in the very-high-resistance films (Al, Pb, and Bi), where it incorrectly predicts a change in temperature dependence in going from the two- to the three-dimensional regime (i.e., when $d/\xi>1$). Since this prediction is so basic to the theoretical picture of fluctuations in superconductors, we are led to postulate one or more of the following explanations: (a) Intrinsic fluctuations go as τ^{-1} in both two and three dimensions. (b) Intrinsic fluctuations do change from τ^{-1} to $\tau^{-1/2}$, but they are of much smaller magnitude than predicted by AL, and the experimental data are dominated by extraneous effects which also go as τ^{-1}. (c) The criterion for observing bulk behavior is not $d/\xi>1$. (d) In highly disordered films the measured normal-state resistance underestimates the mean free path l which enters into the GLAG formula $\xi=0.85(\xi_0 l/\tau)^{1/2}$. This explanation may be tested by a

[34] See K. Maki, in *A Treatise On Superconductivity*, edited by R. Parks (M. Dekker, New York, to be published).

[35] We are indebted to J. J. Hauser for pointing out the relevance of Ref. 33 to the magnetic field effects in thin films.

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[37] A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. **32**, 1442 (1957) [English transl.: Soviet Phys.—JETP **5**, 1174 (1957)].

[38] G. Eilenberger, Phys. Rev. **164**, 628 (1967); K. Maki (unpublished); L. Gruenberg (private communication).

magnetic measurement of ξ via Eq. (15). (See note added in proof.)

In view of the above conclusions, the Bi data are a puzzle: The τ_0 value and the τ^{-1} dependence agree well with the AL predictions, whereas the effects of strong coupling and three-dimensionality are not observed. This circumstance, along wlth the large τ_0 values observed in Al,[4,6] indicates that quantitative discrepancies with the AL theory occur in other materials.

In conclusion, we believe that the present data and analysis cast serious doubt on the validity of previous experimental verification of the AL theory. Until a consistent and verifiable explanation can be given of the various discrepancies discussed above, we must conclude that the previously reported agreement between experiment and theory is, at least in part, fortuitous.

Note added in proof. It has been pointed out to us by Dr. R. S. Thompson that the structure of the films may be such that the mean free path, and therefore the coherence length ξ, is anisotropic. In that case, the ξ relevant for the function G in Eq. (2) might be different from that measured by perpendicular fields [Eq. (15)]. This possibility may be investigated, however, by performing parallel and perpendicular critical field measurements near T_c, and checking the agreement with the GLAG theory.

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Spin Motions in a Classical Ferromagnet*

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The static and dynamic behavior of a simple cubic lattice of classical spins with Heisenberg interactions has been examined by computer on arrays of up to 8192 spins with periodic boundary conditions. Equilibrium values of the energy and magnetization at various temperatures are obtained by Monte Carlo calculations. The results indicate that the magnetization is well approximated by the formula $(1-T/T_c)^\beta$, with $0.32\lesssim\beta\lesssim0.36$ for the full range of temperature from zero to the Curie point T_c. Spin arrays whose energy and magnetization agree with the ensemble averages at a given temperature are taken as characteristic of that temperature. These are then employed to obtain instantaneous and time-displaced spin-correlation functions, the latter involving the numerical solution of the equations of motion of the spin system. The time-displaced correlations show considerable structure. For the most part, spin-wave theory agrees with the low-temperature results. Raising the temperature slows down and smears the structure of the time-displaced correlations. The slowing is much less pronounced than the drop in magnetization. The pulse emanating from a single misaligned spin in a lattice at zero temperature is also calculated.

I. INTRODUCTION

THE statistical mechanical properties of the three-dimensional Heisenberg ferromagnet have been studied by a variety of mathematical techniques, but no exact solutions exist. Some of the properties, such as magnetization and Curie temperature, are better understood than others. In particular, the dynamical behavior of the spin system, except in the spin-wave region, is not known in any detail. Since this dynamical behavior determines the scattering and resonance properties of magnetic materials, it is desirable to obtain a better picture.

Ideally, one would like to study the properties of a system governed by the Hamiltonian

$$\mathcal{H}=-\tfrac{1}{2}\sum_{mn}J(m-n)\mathbf{S}^m\cdot\mathbf{S}^n-g\mu\mathbf{H}\cdot\sum_m\mathbf{S}^m, \qquad (1)$$

where the quantities $\mathbf{S}^m$ are quantum-mechanical spin operators satisfying the usual commutation relations for angular momenta. Such systems, or close approximations thereto, are found in nature, examples being the ferromagnets EuO and EuS, and the antiferromagnet $RbMnF_3$. The Weiss molecular-field approximation[1] provides a qualitative guide to the occurrence of spontaneous magnetization, and series expansion studies and Padé approximant extrapolations[2] based on these expansions yield more refined results for the magnetization. Further, Green's-function analysis[3] has

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[1] See, e.g., C. Kittel, *Introduction to Solid State Physics* (Wiley-Interscience, Inc., New York, 1956), 2nd ed., p. 402.

[2] G. A. Baker, *Advances in Theoretical Physics* (Academic Press Inc., New York, 1965), Vol. 1, p. 1.

[3] D. N. Zubarev, Usp. Fiz. Nauk **71**, 71 (1960) [English transl.: Soviet Phys.—Usp. **3**, 320 (1960)].