Effect of thermodynamic fluctuations on the shape and width of the resistive transition in three-dimensional amorphous superconducting alloys

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Measurements of the fluctuation conductivity above T_c have been made for several bulk amorphous superconducting alloys. The results are compared to existing theories for fluctuation conductivity in three-dimensional superconductors. Agreement with theory is obtained very near T_c, but significant deviations from the predictions of the theory are found for higher temperatures. This is attributed to a breakdown of the Ginzburg-Landau approach. The observed temperature dependence of the fluctuation conductivity was similar in all samples studied and suggests a universal dependence on temperature for bulk amorphous superconductors. The effect of statistical variations in alloy composition (occurring on the scale of the superconducting coherence length ξ) on the width of the superconducting transition widths was observed.

I. INTRODUCTION

Measurements of the enhanced conductivity of a superconductor above T_c were first reported by Glover[1] and explained in terms of evanescent Cooper pairs created by thermal fluctuations. The theoretical work of Aslamasov and Larkin[2] (AL) provided a good account of Glover's results. Later experiments[3-5] and the theoretical work of Maki,[6,7] Thompson,[8] and Patton[9] revealed the importance of another contribution to the fluctuation conductivity in addition to the AL term. A recent review of experimental and theoretical results pertaining to this problem has been given by Craven *et al.*[10] and also by Tinkham.[11] To date most of the experimental work has concerned thin two-dimensional (2D) films obtained by vapor deposition, since fluctuations are most easily observed when one dimension of the sample is small compared to the coherence length of the superconductor. The theoretical work has also emphasized the 2D case.

Conductivity fluctuations in 3D materials are generally difficult to observe as pointed out by several authors.[8,11] The expected magnitude of the AL contribution for a clean 3D superconductor with a coherence length at zero temperature of $\xi(0) \sim 1000$ Å will be of the order of $\Delta\sigma/\sigma_0 \sim 10^{-6}$–$10^{-8}$, where $\Delta\sigma$ is the fluctuation conductivity and σ_0 the normal-state conductivity. The anomalous Maki terms will be of the same order. For an amorphous 3D superconductor, the coherence length can be much smaller and of order $\xi(0) \sim 50$ Å. This is essentially due to the short electron mean free path in an amorphous material. The normal-state conductivity is also reduced by two orders of magnitude. As a result, the AL contribution can be of order $\Delta\sigma/\sigma_0 \sim 10^{-3}$. It should be possible to measure the fluctuation conductivity accurately in this case. Keck and Schmid[12] have recently proposed that strong pair-breaking effects should lead to the disappearance of Maki terms in the fluctuation conductivity of amorphous superconductors. For many amorphous metals, the normal-state conductivity is nearly temperature independent, $1/\sigma_0(d\sigma_0/dT) \lesssim 10^{-4}$, and thus it may easily be subtracted from the total conductivity to give an accurate measure of the temperature dependence of the fluctuation conductivity. The preceding ideas provided the motivation for the present study.

II. EXPERIMENTAL PROCEDURES

An extremely important aspect of measuring fluctuation conductivity concerns the problem of obtaining homogenous samples. Broadening of the superconducting transition resulting from sample inhomogeneity creates difficulty in determining the fluctuation conductivity. Several types of samples were used in this study. These include amorphous $Mo_{30}Re_{70}$ and $Nb_{75}Ge_{25}$ samples prepared by rf sputtering with thicknesses of 0.2–1 μm, and also amorphous $La_{78}Au_{22}$ (Ref. 13) and $La_{75}Au_{15}Cu_{10}$ samples prepared by liquid quenching in a vacuum of $\sim 10^{-4}$ Torr. The latter samples are in the form of foils with thickness ~ 50 μm. The ternary composition was chosen to give a nearly temperature-independent normal-state conductivity, $1/\sigma_0(d\sigma_0/dT) < 10^{-5}$. The structure of the samples used was checked by x-ray diffraction. Samples used for the conductivity measurements were those showing no indication of crystallization in the x-ray scans.

Resistivity measurements were made using a standard four-probe technique. Typical current densities of $\lesssim 1$ A/cm^2 were used in the measurements. The temperature was measured using a

TABLE I. List of composition, method of preparation, transition temperature, transition width, excess conductivity at $2T_c$, and residual resistance for the alloys used in this study.

Alloy	Method of preparation	T_c (°K)	ΔT_c (°K)	$[(\Delta\sigma)_{2T_c}]/\sigma_0$	ρ_0 ($\mu\Omega$cm)
$La_{78}Au_{22}$	Liquid quenching	3.934	0.018	0.000 32	280 ± 60
$La_{78}Au_{22}$ [a]	Liquid quenching	3.765	0.055	0.000 22	260 ± 60
$La_{75}Au_{15}Cu_{10}$	Liquid quenching	3.795	0.022	0.000 35	320 ± 60
$Mo_{30}Re_{70}$	rf sputtering	7.620	0.021	0.000 66	...
Nb_3Ge	rf sputtering	3.342	0.014	0.000 74	...

[a] With evidence of crystallization.

germanium resistance thermometer with an absolute accuracy of 0.1°K and a relative accuracy of 0.001°K for temperatures in the vicinity of the superconducting transitions. The resistance was measured with an accuracy of better than one part in 10^4.

III. RESULTS AND ANALYSIS

The measured superconducting transition temperatures of the samples studied are given in Table I along with the composition and method of preparation for each alloy. The transition temperature given was taken to be the highest temperature for which the measured resistance was zero. The width of the transitions as indicated by difference between the temperatures at which the resistance was 25% and 75% of its normal-state value is also given in Table I. Several samples were measured for each of the alloy compositions listed in the table, and the results presented represent those for the samples having the sharpest transition. One $La_{78}Au_{22}$ sample showing evidence of crystallization in the x-ray-diffraction scan is also listed for comparison in the table. Comparing this to a more homogeneous $La_{78}Au_{22}$ sample showing only the amorphous structure shows that a significant increase in the transition width occurs with the onset of crystallization. This can presumably be attributed to inhomogeneity resulting from phase segregation in the crystallization process.

The normalized resistance $R(T)/R_0$ (with R_0 the normal-state resistance) as a function of temperature for amorphous $La_{75}Au_{25}Cu_{10}$ and $Mo_{30}Re_{70}$ samples are shown in Fig. 1. As previously mentioned, the normal-state resistance for the $La_{75}Au_{15}Cu_{10}$ sample is nearly temperature independent with $[1/\sigma_0(d\sigma/dT)] \sim 10^{-5}$ for temperatures above $\sim 3T_c$. The uniform nature of the liquid quenched foils permitted a reasonably accurate determination of the value of ρ_0. These values are listed in Table I for reference. Since T_c was defined by $R(T_c) = 0$, it can be seen in Fig. 1 that no significant rounding of the resistive transition is observed on the low-temperature side of the transition. Such rounding would be expected if significant inhomogeneity were present in the samples and thus its absence provides evidence of homogeneity.

The rounding of the transitions above T_c is visible up to temperatures exceeding $2T_c$ in Fig. 1. At $2T_c$, the normalized excess conductivity $\Delta\sigma/\sigma_0$ is of order 10^{-3}. Values of $\Delta\sigma/\sigma_0$ at $2T_c$ for all samples studied are given in Table I for comparison. The values of $\Delta\sigma/\sigma_0$ at $2T_c$ range from 3.2×10^{-4} for $La_{78}Au_{22}$ to 7.4×10^{-4} for the Nb_3Ge alloy. The resistive transitions for all samples are similar to those shown in Fig. 1. The insert in Fig. 1 shows a magnified portion of the resistivity curve for amorphous $Mo_{30}Re_{70}$.

The temperature dependence of the excess conductivity is more clearly illustrated in Fig. 2. The figure shows $\ln(\Delta\sigma/\sigma_0)$ as a function of $\ln t$ where t is the reduced temperature $t = (T - T_c)/T_c$ for $Mo_{30}Re_{70}$, $La_{85}Au_{15}Cu_{10}$, $La_{78}Au_{22}$, and the $La_{78}Au_{22}$ sample showing evidence of crystallization previously mentioned. Several features of the behavior of the excess conductivity can be observed. First, discontinuous change in the temperature dependence of the fluctuation conductivity near

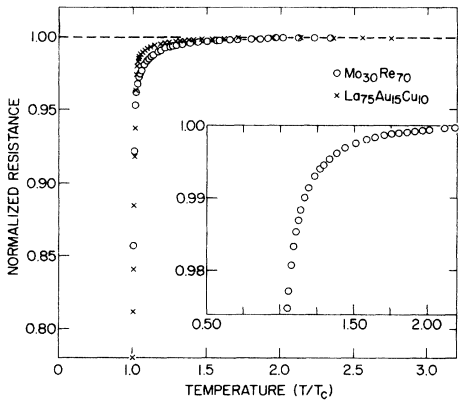

FIG. 1. Normalized resistance as a function of temperature (in units of T_c) for amorphous $Mo_{30}Re_{70}$ and amorphous $La_{75}Au_{15}Cu_{10}$.

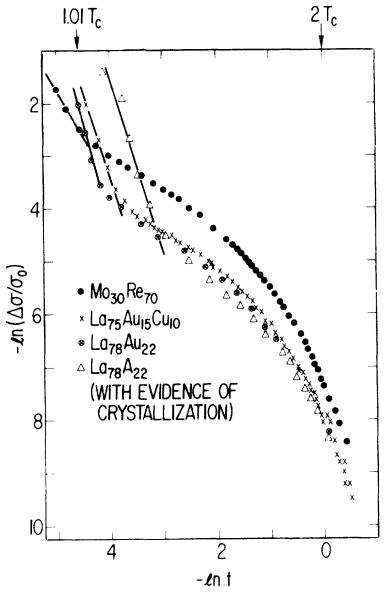

FIG. 2. Logarithmic plot of the normalized excess conductivity $(\Delta\sigma/\sigma_0)$ as a function of the reduced temperature $t=(T-T_c)/T_c$. Samples include amorphous $Mo_{30}Re_{70}$, $La_{75}Au_{15}Cu_{10}$, $La_{78}Au_{22}$, and the $La_{78}Au_{22}$ sample showing evidence of crystallization. This sample was prepared under identical conditions as the amorphous sample but with lower cooling rate.

T_c ($t \lesssim 0.02$) may be attributed to sample imhomogeneity. For the sample showing evidence of crystallization, this effect extends over a larger temperature range $t \lesssim 0.05$ showing this sample to be less homogeneous than the corresponding amorphous sample prepared under identical conditions. This will be discussed in more detail in a later section. It should be noted that the exact temperature range for which transition broadening is significant is somewhat arbitrarily defined. Adjusting the experimental value of T_c yields large relative changes in the resulting value of t (and ln t) when $t \to 0$. We have defined T_c by the $R=0$ criteria. Previous work[14] suggests that less than half of the sample must be superconducting in order to establish the continuity of superconducting regions required to give $R=0$. On this basis, one expects the mean T_c of the sample to be near but slightly lower than T_c as defined by the $R=0$ criteria. Inspection of Fig. 2 shows that if slightly lower T_c is chosen, the result will be a small increase in the apparent transition-broadened region. A choice of higher T_c would reduce the apparent region somewhat but does not seem very reasonable on the basis of the above argument. The width of broadened transition region will be discussed in more detail in Sec. IV. It should be pointed out that T_c has been used as an adjustable parameter in fitting experimental data to theoretical expressions for the fluctuation conductivity by many investigators. This procedure is rather arbitrary and can lead to spurious agreement between theory and experiment very close to T_c. In the present study, the comparison with theory will not emphasize the region very close to T_c ($t \lesssim 0.02$).

Considering the temperature range far enough away from T_c to avoid the problem discussed above, one finds that the temperature dependence of the fluctuation conductivity is very similar for all samples studied up to the highest temperatures for which the fluctuation conductivity is measurable ($\sim 3T_c$). The data for the amorphous $La_{78}Au_{22}$ and Nb_3Ge samples are not shown in Fig. 2 to avoid overlap of the data points with those presented. The detailed temperature dependence of the fluctuation conductivity will be discussed in Sec. IV and compared to theory.

IV. DISCUSSION

A. Theoretical background

The fluctuation conductivity of a 3D superconductor above T_c can be written as the sum of the two contributions

$$\Delta\sigma = \sigma_{AL} + \sigma_{MT}, \tag{1}$$

where σ_{AL} is the contribution originally discussed by Aslamasov and Larkin[2] and σ_{MT} is the contribution of the anomalous or Maki-Thompson terms.[6-8] The second term has been found to be essential in describing the anomalously large fluctuation conductivity of thin films with small pair-breaking effects.[10] In the 3D case, the AL term can be written[8]

$$\sigma_{AL}(3D) = e^2/32\xi t^{1/2}, \tag{2}$$

where ξ is the coherence length at zero temperature and may be obtained from measurement of the temperature dependence of H_{c2} using the expression $H_{c2} = |t|/2e\xi^2$. In practical units, $e^2 = 2.43 \times 10^{-4}\Omega^{-1}$. The reduced temperature t is the same as that defined previously but changes sign below T_c. In the absence of pair breaking, the Maki term for the 3D case is simply

$$\delta_{MT}(3D) = e^2/8\xi t^{1/2} \tag{3}$$

which is just $4\sigma_{AL}(3D)$. In the presence of pair breaking, the Maki term becomes[15]

$$\sigma_{MT}(3D) = (e^2/8\xi t^{1/2})\{1/[1+(\delta/\epsilon_0)^{1/2}]\}, \tag{4}$$

where $\epsilon_0 = 8/\tau(T-T_c)$ and δ represents the pair-breaking parameter. For large δ, $\sigma_{MT} \to 0$. A similar result for the 2D case was used to explain the absence of Maki terms in the fluctuation conductivity of amorphous Bi films and lead films for which agreement was found with the simple AL

theory.[10,11] It was argued that δ should be large for these materials, and thus that Maki terms should be small.

Appel[16] has shown that the pair-breaking parameter can be generally related to other material parameters through the following expression:

$$\delta \propto \lambda / M\Theta_D^2 \tag{5}$$

where λ is the electron-phonon coupling constant, M is the atomic mass, and Θ_D is the Debye temperature. Keck and Schmid[12] have pointed out that in amorphous materials λ is generally large, and Θ_D^2 small. Using a detailed argument involving the Eliashberg function $\alpha^2F(\omega)$, they showed that the pair-breaking parameter will be very large and thus should generally lead to suppression of Maki terms for amorphous superconductors. Indeed, the results for 2D amorphous films support this argument.[10]

Finally, it should be pointed out that the theoretical calculations of the fluctuation conductivity in the literature[2,6-9] concern the temperature range near T_c. The use of the Ginzburg-Landau free-energy functional involves the assumption that not only ψ, but also its derivatives vary slowly in space. When $T \sim 2T_c$, for example, $\xi(t)$ becomes less than $\xi(0)$ and short-wavelength fluctuations become increasingly important. The slow-variation approximation certainly breaks down in this case. This problem has been discussed by Gollub *et al.* in conjunction with the behavior of the fluctuation diamagnetism.[17] Experimentally, it was found that large deviations from Ginzburg-Landau–type theories occur in the fluctuation diamagnetism of 3D samples[17,18] for temperatures far from T_c.

B. Comparison of results with theory

Before attempting a comparison of experimental results for the fluctuation conductivity with theory, it is worthwhile to digress briefly on the problem of transition broadening in somewhat more detail. It has been experimentally observed (see Table I and Fig. 2) that the width of the broadened transition region is of the order of 0.02 °K in all of the amorphous alloys studied. Since several amorphous samples were used, and two very different preparation techniques employed, it seems worthwhile to consider the origin of this broadening.

In addition to temperature, there are other thermodynamic variables for which fluctuations about the mean values may be important. These include alloy composition and atomic density to name two. As in the case of temperature fluctuations, the fluctuation in these parameters become important when the characteristic volume considered becomes small. A detailed discussion of composition and density fluctuations is given by Landau and Lifshitz.[19] In the present case we take the characteristic volume to be determined by the superconducting coherence length at zero temperature ξ. Using the volume $V \approx \frac{4}{3}\pi\xi^3$, and designating the mean atomic volume as v_a, one finds that the characteristic volume contains V/v_a atoms. For a binary alloy with composition $A_{1-X}B_X$, there are on the average $(1-X)N$ A atoms and XN B atoms in this volume. If the alloy is taken to be an ideal random solid solution, then the root-mean-square deviation in the number of B atoms will be roughly $(NX)^{1/2}$ giving a mean deviation of $\langle \Delta X^2 \rangle^{1/2} \approx X/(NX)^{1/2}$ in the compositional variable X. For an amorphous superconducting alloy such as $La_{78}Au_{22}$, $\xi \sim 70$ Å.[13] This gives $N \sim 10^4$ atoms in a characteristic volume and $\langle \Delta X^2 \rangle^{1/2} \sim 10^{-2}X$. The derivative of T_c with respect to composition for the La-Au system can be taken from Ref. 13 and is $dT_c/dX \approx 0.05$ °K/at.% yielding an expected value of

$$\langle \Delta T_c^{\,2} \rangle = \frac{dT_c}{dX} \langle \Delta X^2 \rangle^{1/2} \approx 0.01\ ^\circ\mathrm{K} \tag{6}$$

for the broadening of the transition temperature of this amorphous alloy. The main assumption made is that a volume with characteristic dimension ξ will exhibit a transition temperature characteristic of the composition within that volume. This assumption can be put on a firm basis by appealing to the theory of the proximity effect. Another argument similar to that above can be made which shows that broadening of the transition due to fluctuations in atomic density will yield broadening effects of nearly the same magnitude. Density fluctuations can be interpreted in terms of a variation in the effective hydrostatic pressure in different regions of the sample which broaden the transition through the dependence of T_c on pressure (dT_c/dp). This dependence is quite large for lanthanum.[20]

The observed magnitude of transition broadening for the La-base alloys is indeed on the order predicted by the above arguments. All samples studied show broadening effects of this magnitude. The preceding argument suggests that the observed broadening can be explained as a general property of amorphous superconductors which results from fluctuations in fundamental thermodynamic variables. This has interesting implications for the study of critical phenomena in amorphous superconductors and more generally for any superconductor with small coherence length. For the present study, the preceding implies that the fluctuation conductivity may be generally inaccessible near T_c in amorphous superconductors. With this in mind, we return to the problem of comparing the present results to theory.

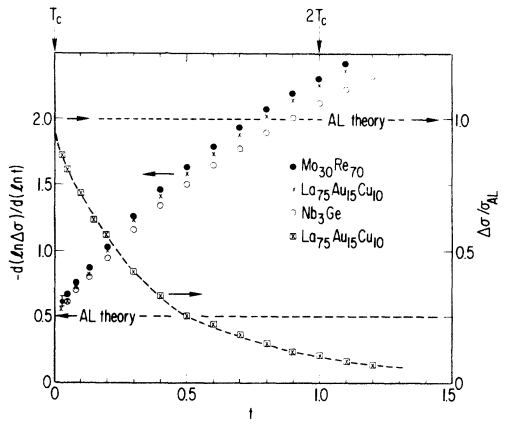

FIG. 3. Comparison of the temperature dependence and magnitude of the excess conductivity with the predictions of the AL theory for several amorphous samples. Values corresponding to the AL theory are indicated by the dashed lines.

On the basis of the suggestion by Keck and Schmid[12] that large pair-breaking effects should suppress the Maki terms in the fluctuation conductivity for amorphous materials, we begin by calculating the theoretical value of the AL term for the samples studied. This requires a knowledge of $\xi(0)$, the zero-temperature coherence length, as determined from a measurement of $H_{c2}(T)$. These data for $La_{78}Au_{22}$ are taken from Ref. 13. Linear extrapolation gives $H_{c2}(0) \approx 80$ kG which in turn gives $\xi = 63$ Å. One finds for $La_{78}Au_{22}$

$$\sigma_{AL} = (12.1/t^{1/2})\ (\Omega\,\mathrm{cm})^{-1}.$$

Since $\sigma_0 = 1/\rho_0$ and using $\rho_0 = 280\ \mu\Omega$ cm for $La_{78}Au_{22}$, one obtains

$$\sigma_{AL}/\sigma_0 = 3.37 \times 10^{-3}/t^{1/2}. \tag{7}$$

This may be compared to the experimental results by plotting the ratio $r = \Delta\sigma/\sigma_{AL}$ as a function of t. If the AL term alone describes the fluctuation conductivity, then this ratio should be 1 for all t. Only data lying outside the transition-broadened region (shown in Fig. 2) were used in this comparison in order to avoid over estimating the experimental fluctuation conductivity. The result is shown in Fig. 3. It can be seen that in the limit $T \to T_c$ $(t \to 0)$ the value of $r \approx 1$. The error in the calculation value of σ_{AL} is about $\pm 10\%$ and comes from the uncertainty in $H_{c2}(0)$. The error in $\Delta\sigma$ is approximately $\pm 20\%$ coming mainly from the uncertainty in $\rho_0(\sigma_0)$. The error in the ratio $\Delta\sigma/\sigma_{AL}$ is thus $\sim \pm 25\%$. Thus within the error we find that in the limit $t \to 0$, the observed fluctuation conductivity is accounted for entirely by the AL contribution.

For the amorphous Nb_3Ge, data for $H_{c2}(T)$ are not available. However, for high disordered Nb_3Ge films with T_c range from 7.5 to 17 °K it was found that $(dH_{c2}/dT)_{T=T_c} \approx 100$ kG/°K and $H_{c2}(0) \approx 350$ kG independent of T_c.[21] On this basis, we estimate $H_{c2}(0) \approx 300$ kG for the amorphous Nb_3Ge. This in turn gives a coherence length $\xi(0) \sim 30$ Å. The theoretical value of σ_{AL} should be about twice that of the La-Au alloys. The observed values of $\Delta\sigma/\sigma_0$ for Nb_3Ge are also twice those observed for $La_{78}Au_{22}$ (see Table I) and therefore it can be concluded that the fluctuation conductivity for Nb_3Ge may also be explained in terms of an AL term in the limit $t \to 0$. An estimate of $H_{c2}(0)$ for amorphous $Mo_{30}Re_{70}$ based on measurements for similar binary amorphous alloys containing only transition metals[22] gives $H_{c2}(0) \sim 300$ kG. Roughly the same value of ξ and σ_{AL} are estimated as for Nb_3Ge and again the observed values of $\Delta\sigma$ are twice those of $La_{78}Au_{22}$. Summarizing, the observed magnitude of the fluctuation conductivity near T_c $(t \to 0)$ can be accounted for by the AL term alone in all samples. In the case of the lanthanum alloys, all parameters required to calculate σ_{AL} were taken from direct measurement.

In order to compare the data with the temperature dependence predicted by the AL theory the slope $d(\ln\Delta\sigma)/d(\ln t)$ was plotted as a function of t. For the AL theory $d[\ln\sigma_{AL}(3D)]/d(\ln t) = 0.5$ [see Eq. (2)] for all t. In Fig. 3, this is compared to the data for all three alloys studied. Again, data in the broadened transition region were not used. For $t \to 0$, it can be seen that $d(\ln\Delta\sigma)/d(\ln t) \to 0.5 \pm 0.05$ for all three materials. We find good agreement with the AL theory in the region near T_c.

It can be concluded from the preceding that the AL term alone can account for both the temperature dependence and the magnitude of the observed flcutuation conductivity near T_c. Combined with the argument of Keck and Schmid,[12] this provides strong evidence for the general absence of Maki terms in the case of 3D amorphous superconductors. The AL theory does not give satisfactory agreement with the data for $t \gtrsim 0.05$. At high temperatures $(t \sim 1)$, the disagreement is large. The magnitude of $\Delta\sigma$ falls well below the AL prediction at high t and the temperature dependence of $\Delta\sigma$ is not describable by any power law. A good fit to the temperature dependence of $\Delta\sigma$ is illustrated in Fig. 4 for $La_{75}Au_{15}Cu_{10}$ and $Mo_{30}Re_{70}$. It can be seen that $\Delta\sigma \propto e^{-\gamma t^{1/2}}$ over a wide range of t where γ is approximately 4.5 for all samples studied. Several exponential functions were used to fit the data with the above expression giving the best result for a broad range of t. Since the above function is nonsingular for $t \to 0$, it is clear that it cannot describe the observed behavior near T_c. The data in the region near T_c deviate from this form as can

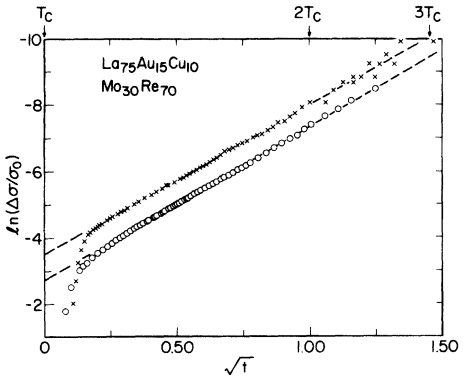

FIG. 4. The logarithm of the normalized excess conductivity as a function of $\sqrt{t}$ for two amorphous samples showing the exponential dependence of the fluctuation conductivity on temperature over a range of several orders of magnitude.

be seen in Fig. 4.

The apparent breakdown of the AL theory for $t \gtrsim 0.05$ can be interpreted as resulting from the use of the Ginzberg-Landau free-energy functional. The slow variations approximation discussed in Sec. IV A should be expected to fail for large t. The increased importance of short-wavelength fluctuations must take into account at higher temperature.[17] Perhaps the most surprising result of the present work is that the failure of the theory occurs for quite small values of $t(\sim 0.05)$. This may signal the need to go beyond the mean-field type of theory in accounting for critical behavior of dirty 3D superconductors. It is interesting that in the 2D case, evidence for failure of the mean-field theory was not found even for quite large t values.[10]

Finally, the present results can be compared to that of Gollub *et al.* on the fluctuation diamagnetism of 3D superconductors.[17,18] Large deviations from theoretical predictions based on the Ginzberg-Landau free-energy functional were also observed in that work. The high-temperature fluctuation diamagnetism was found to have a roughly exponential temperature dependence. This result is in qualitative agreement with the present work and strengthens the interpretation of the results found here.

V. CONCLUSIONS

Measurements of the fluctuation conductivity of several three-dimensional (bulk) amorphous superconducting alloys have been made. The temperature dependence of the fluctuation conductivity was found to be nearly identical in all materials studied. It is thus proposed that this dependence is characteristic of three-dimensional amorphous superconductors. The theoretical predictions of the Aslamasov-Larkin theory were found to provide a good quantitative account of the data near T_c. Evidence was presented to show that anomalous or Maki-Thompson terms are not present in the fluctuation conductivity. For higher temperatures ($t \gtrsim 0.05$), the predictions of the Aslamasov-Larkin theory are found to disagree significantly with experimental results. It is suggested that this is a result of the breakdown of the Ginzberg-Landau free-energy functional.

The importance of transition broadening in amorphous superconductors was discussed in some detail. It was suggested that compositional and atomic-density fluctuations of a thermodynamic origin limit the width of the superconducting transition in amorphous alloys. Estimates of transition broadening based on a simple thermodynamic argument give broadening effects of the order observed. The present results suggest a need for additional theoretical work aimed at extending the description of fluctuation conductivity to temperatures beyond the region of validity of the mean-field theory.

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