

## Superconducting fluctuation effects in the resistive transition of amorphous bismuth films\*

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Improved measurements were made of the temperature-dependent resistivity of amorphous Bi films in order to observe the effects of superconducting fluctuations. Measurements were made in the temperature range  $T_c \leq T \leq 2.5T_c$  on films ranging from 500 to 5000 Å thick. Temperature resolution near  $T_c$  of 0.2 mK and precise resistance measurements at higher temperatures (1 part in  $10^4$ ) permitted a detailed comparison with theory. Films over 500 Å thick showed a temperature-dependent transition from thin-film (two-dimensional) behavior to bulk (three-dimensional) behavior in agreement with the predictions of Aslamzov and Larkin.

### I. INTRODUCTION

During the past several years numerous studies have been made of superconducting fluctuation rounding of the resistive transition of thin-film superconductors.<sup>1,2</sup> The usual procedure in analyzing resistive transition data has been to perform a multiparameter least-squares fit to a theoretical prediction for the fluctuation conductivity. A difficulty with this procedure is that adjustment of the theoretical parameters will generally permit the Aslamzov-Larkin (AL) theory<sup>3</sup> or other<sup>4-7</sup> theories of the resistive transition to fit any smooth data over at least a portion of the transition region. Testardi *et al.*<sup>8</sup> have pointed out that a more critical test of proposed theories would be an experimental observation of the predicted change in the temperature dependence of the resistance corresponding to the transition between the two- and three-dimensional fluctuation regimes. Several investigators have attempted to study the two- to three-dimensional transition; however, in most cases there have been some doubts about the results. Testardi *et al.*<sup>8</sup> found only two-dimensional behavior in Pb films that were supposedly in the three-dimensional regime. Gittleman *et al.*<sup>9</sup> observed the  $\tau^{-1/2}$  (where  $\tau = \ln T/T_c$ ) temperature dependence predicted by the AL theory for fluctuations in the three-dimensional regime. However, their measurements on granular Al and Sn films did not show a two- to three-dimensional transition. Goldman *et al.*<sup>10</sup> believe they have observed the transition in  $\text{Nb}_{0.88}\text{Ti}_{0.12}\text{N}$  films, however, their results were highly dependent on the method used to analyze the data. Inspection of their Fig. 1 suggests that the films they used were not uniform; furthermore, the films proved to have a temperature-dependent normal-state resistance which greatly complicated the data analysis. Sixl and Sanwald<sup>11</sup> claim to observe zero, one, two, and three-dimensional behavior in quench-condensed Al films, but their

results do not show the two- to three-dimensional transition. In addition, Granqvist<sup>12</sup> has shown that their results can be interpreted as due to a thickness-dependent pair-breaking parameter. Finally, Johnson and Tsuei<sup>13</sup> have made measurements on a variety of bulk amorphous alloys. Very close to  $T_c$  they observe a  $\tau^{-1/2}$  temperature dependence; however, they do not observe the transition to the two-dimensional regime, perhaps because of transition broadening due to statistical variations in alloy composition. The best observations of the two- to three-dimensional transition are Glover and Naugle's work<sup>14</sup> on ~3000 Å thick amorphous Bi films. However, additional measurements with higher accuracy and covering a larger range of film thicknesses offer the possibility of a more definitive test of proposed theories.

The measurements reported here were made on amorphous Bi films with work concentrated in the temperature range where mean-field theories such as the AL theory<sup>3</sup> are expected to apply. Film thicknesses ranged from 500 to 5000 Å. The measurements covered the temperature range  $T_c \leq T \leq 2.5T_c$ . Special attention has been given to a discussion of the analysis procedure.

### II. THEORETICAL CONSIDERATIONS

Assuming that normal-state and fluctuation conductivity occur in parallel, the AL theory gives the following expression for the temperature-dependent resistance of a film of thickness  $D$ <sup>3,15</sup>:

$$\frac{R_0 - R(T)}{R(T)} = \frac{e^2}{16\hbar} \frac{1}{\sigma_0 D \tau} G(\tau) = \frac{\sigma'_{\text{AL}}}{\sigma_0}, \quad (1)$$

$$G(\tau) = \frac{1}{2} \{ 1 + D/\xi_T \coth D/\xi_T \}, \quad (2)$$

$$\xi_T = \eta/\tau^{1/2}, \quad (3)$$

where  $R_0$  is the normal-state residual resistance,  $R(T)$  is the temperature-dependent resistance of the film,  $\sigma'_{\text{AL}}$  is the fluctuation conductivity,  $\sigma_0$

is the residual normal-state conductivity,  $\tau = \ln T/T_c^{mf}$ , and  $\eta$  is the Ginzburg-Landau coherence length constant. The notation  $T_c^{mf}$  is used to indicate that the transition temperature is to be considered a theoretical parameter; thus, the film resistance will not necessarily be zero at  $T_c^{mf}$ .

In the two-dimensional limit  $D \ll \xi_T$ , the AL theory becomes

$$\frac{R_0 - R(T)}{R(T)} = \tau_0^{AL} / \tau, \quad (4)$$

while in the three-dimensional limit  $D \gg \xi_T$ , the AL theory becomes

$$[R_0 - R(T)]/R(T) = \frac{\tau_0^{AL}}{2\tau^{1/2}} \frac{D}{\eta}, \quad (5)$$

where  $\tau_0^{AL} = (e^2/16\hbar)R_{0\Box}$  with  $R_{0\Box} = 1/\sigma_0 D$ .

According to the AL theory, whether a sample exhibits two-dimensional or three-dimensional behavior depends only on the ratio of the sample thickness  $D$  to the Ginzburg-Landau coherence length  $\xi_T$ . Since  $\xi_T$  decreases with increasing temperature, the AL theory predicts a change from a  $\tau^{-1}$  temperature dependence characteristic of two-dimensional fluctuations to a  $\tau^{-1/2}$  dependence characteristic of three-dimensional fluctuations when the temperature is raised sufficiently far above  $T_c^{mf}$ .

To properly test the AL theory, measurements must be made on a single thin film in *both* the two-dimensional ( $\xi_T \gg D$ ) and the three-dimensional ( $\xi_T \ll D$ ) regimes. As Goldman *et al.*<sup>10</sup> emphasize, the measurements must be made on a well-characterized material and the analysis should be done in such a way as to avoid the problems inherent in multiparameter fits. Furthermore, measurements made on films of different thickness should all show comparable behavior with a systematic dependence on film thickness.

### III. EXPERIMENTAL DETAILS

A schematic of the cryostat-evaporator used for the experiment is shown in Fig. 1. The vacuum in the vicinity of the sample was  $\sim 10^{-8}$  Torr. Z-cut, polished, crystalline quartz substrates were mounted on a substrate holder attached to the innermost liquid He tank. A germanium resistance thermometer was mounted on the rear of the substrate holder along with a small heater used to raise the temperature above 4.2 K. A mask mounted on the front of the substrate holder  $\sim 75$   $\mu\text{m}$  above the surface of the substrate was used to define the film geometry.

Three films were produced simultaneously by depositing Bi(Tl) onto the quartz substrate; during deposition the temperature of the substrate rose

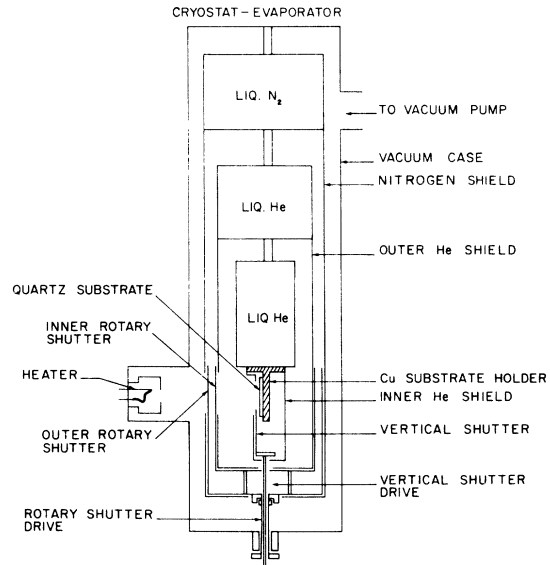


FIG. 1. Cryostat evaporator. Bismuth evaporated from a crucible held in the heater was deposited on the quartz substrate which was held at cryogenic temperatures by the lower of the two helium reservoirs.

to between 7 and 10 K. The purpose of the Tl was to stabilize the amorphous phase<sup>16,17</sup> which otherwise tended to crystallize at  $\sim 12$  K. With 3 at. % Tl the films were stable up to  $\sim 18$  K; additional Tl raised the crystallization temperature as high as 100 K. The Bi(Tl) mixtures were prepared by the method described by Shier and Ginzberg.<sup>18</sup>

The Bi(Tl) mixtures were evaporated from a resistively heated Mo crucible with an 0.6-mm orifice. The crucible was located  $\sim 30$  cm from the substrate. From geometrical considerations alone the thickness uniformity of the films is  $\sim \frac{1}{2}\%$ . The shadowed "edge" regions of the films are computed to be  $\sim 0.2$   $\mu\text{m}$  wide.

The temperature was determined with a germanium resistance thermometer calibrated by the National Bureau of Standards to an absolute accuracy of  $\pm 20$  mK. The thermometer resistance was plotted on an X-Y recorder or read from a digital millivoltmeter. The film resistance was determined using a four-terminal potentiometric method. A mercury cell warmed to  $40^\circ\text{C}$  supplied a current constant to  $\sim 1/50000$  per hour. The measurements were made with a current of 100  $\mu\text{A}$ ; reducing the current to 10  $\mu\text{A}$  did not change the results within experimental error. The voltage drop across the film was plotted on an X-Y plotter in the temperature interval  $T_c \leq T \leq T_c + 0.1$  K where the resistance changes rapidly with temperature. At higher temperatures the voltage drop was determined with a microvolt potentiometer with a resolution of 1 part in  $10^4$ . A summary of the

TABLE I. Measurement errors.

	$T_c < T < T_c + 0.1$ K	$T > T_c + 0.1$ K
Film resistance	$\pm 0.15\% R_0$	$\pm 0.004 \Omega$
Relative temperature	$\pm 0.2$ mK	$\pm 0.8$ mK at $\sim 6$ K increasing to $\pm 50$ mK at $\sim 18$ K
Absolute temperature	$\pm 20$ mK	$\pm 20$ mK

errors involved in the measurements is given in Table I. Further discussion of the experimental details can be found elsewhere.<sup>19</sup>

#### IV. RESULTS AND ANALYSIS PROCEDURE

The data consists of resistance vs temperature measurements for thirteen amorphous Bi films.<sup>19</sup> The procedure adopted to analyze the data is outlined below. (i) The normal-state residual resistance  $R_0$  was determined from the relation

$$R_0 \equiv 1.0025R_{\max} \quad (6)$$

where  $R_{\max}$  was the maximum value of resistance measured for the film being analyzed. Generally,  $R_{\max}$  occurred at  $T \approx 12$  K (Fig. 2). (ii) Assuming that the AL theory correctly describes the data, and using the fact that close to  $T_c$ ,  $\ln T/T_c \approx (T - T_c)/T_c$ , Eq. (4) shows that a plot of  $R(T)/[R_0 - R(T)]$  vs  $T$  will be linear with a slope equal to  $(T_c^{mf} \tau_0)^{-1}$  and an extrapolated intercept at  $T = T_c^{mf}$  (Fig. 3). The notation  $\tau_0$  indicates the experimentally determined value of the AL parameter  $\tau_0^{AL}$ . The uncertainty in  $\tau_0$  and  $T_c^{mf}$  ranges from  $\pm 4\%$  and  $\pm 0.4$  mK, respectively for the thinnest films, to  $\pm 10\%$  and  $\pm 0.2$  mK for the thickest films. The dominant source of error in  $\tau_0$  and  $T_c^{mf}$  is the accuracy of the resistance measurement. Error in

the determination of  $R_0$  has an insignificant effect on the values of  $T_c^{mf}$  and  $\tau_0$ . (iii) The film thickness  $D$  was determined from the relation

$$D = \rho_0/R_{0\Box}, \quad (7)$$

with  $\rho_0 = 155 \mu\Omega \text{ cm}$ .<sup>20</sup> The thickness of seven amorphous Bi + 3 at. % Tl films made in conjunction with this study was determined by the Tolansky method; the measured thickness agreed with those calculated from Eq. (7) to  $\pm 10\%$ . (iv) After determining  $R_0$ ,  $T_c^{mf}$ ,  $\tau_0$ , and  $D$  as described above, the Ginzburg-Landau coherence length constant  $\eta$  was determined by fitting Eq. (1) to the data at  $\tau = 0.050$ . This point was chosen because it is well into the three-dimensional fluctuation region for all but the thinnest films, yet it is still below the temperature at which variation in the normal-state resistance affects the measurements. The uncertainty in  $\eta$  is almost entirely due to uncertainty in the determination of  $R_0$ . A change in  $R_0$  of 0.1% produces approximately a 15% change in  $\eta$ . For films over 1000 Å thick the uncertainty in  $\eta$  is estimated to be  $\pm 20\%$ . Because finite thickness effects are smaller in the thinner films at  $\tau = 0.050$ , the uncertainty in  $\eta$  is correspondingly larger. For the three films which are less than 1000 Å thick the uncertainty in  $\eta$  is  $\sim 30\%$ .

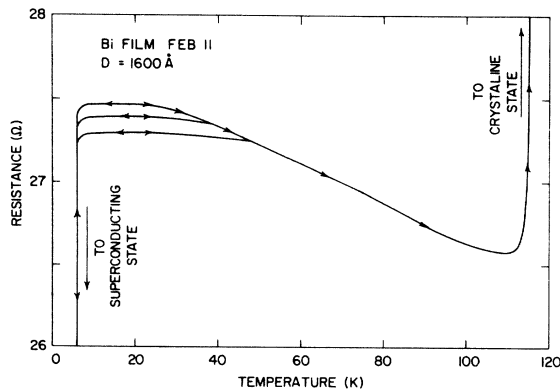


FIG. 2. Annealing curve of an amorphous bismuth film. The double-ended arrows ( $\leftrightarrow$ ) indicate "reversible" behavior. The single-ended arrows ( $\rightarrow$ ) indicate "irreversible" behavior.

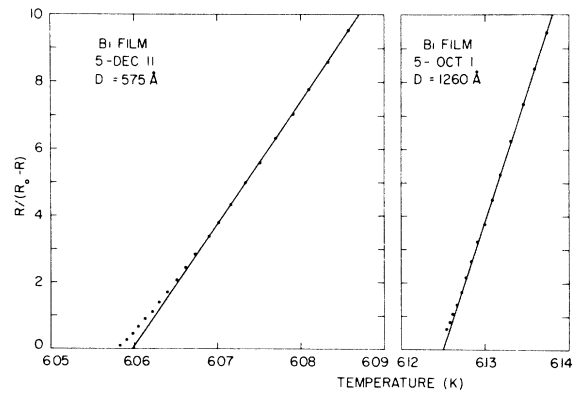


FIG. 3.  $R/(R_0 - R)$  as a function of temperature for two amorphous Bi films. The straight line drawn through the data points was used to determine the transition temperature  $T_c^{mf}$  and the experimental transition width  $\tau_0 T_c^{mf}$ . Error bars are the size of the plotted points.

TABLE II. Experimental parameters determined from comparison with the Aslamazov-Larkin theory.

Film No.	$R_{0\Box}(\Omega)$	$\frac{\tau_0}{R_{0\Box}}(\Omega^{-1})$	$T_c^{mf}$ (K)	$\eta(\text{\AA})$	$D(\text{\AA})$	$\frac{\text{Film length}}{\text{Film width}}$
1-Jun 24	32.335	$1.66 \times 10^{-5}$	6.0449	86	480	5.38
2-Jun 24	32.342	$1.66 \times 10^{-5}$	6.0452	86	480	5.54
1-Aug 23 <sup>a</sup>	5.440	$1.74 \times 10^{-5}$	6.1604	103	2850	5.38
2-Aug 23 <sup>a</sup>	5.359	$1.77 \times 10^{-5}$	6.1606	103	2890	5.54
1-Aug 31	9.636	$1.65 \times 10^{-5}$	6.1443	99	1610	5.38
2-Aug 31	9.517	$1.67 \times 10^{-5}$	6.1448	99	1630	5.54
1-Sep 10	8.166	$1.66 \times 10^{-5}$	6.1391	101	1900	5.38
2-Sep 10	8.078	$1.67 \times 10^{-5}$	6.1397	100	1920	5.54
5-Oct 1	12.357	$1.68 \times 10^{-5}$	6.1251	99	1255	5.44
4-Oct 18	5.564	$1.70 \times 10^{-5}$	6.1350	100	2785	5.14
5-Oct 18	5.563	$1.70 \times 10^{-5}$	6.1343	98	2785	5.44
5-Dec 3 <sup>a</sup>	3.119	$1.78 \times 10^{-5}$	6.1643	99	4970	5.44
5-Dec 11	27.049	$1.62 \times 10^{-5}$	6.0599	85	575	5.44

<sup>a</sup>Bi + 6 At. % Tl.

The data for several of the films is shown in Fig. 4. The values of  $R_{0\Box}$ ,  $\tau_0/R_{0\Box}$ ,  $T_c^{mf}$ ,  $\eta$  and  $D$  are given in Table II for thirteen amorphous Bi films including those for which the data is plotted in Fig. 4.

#### V. NORMAL-STATE RESISTANCE

Since the fluctuation conductivity is so small at higher temperatures, it is important to have an accurate determination of the residual resistance  $R_0$ . For this reason, special attention was paid to the normal-state properties of the films.

In the vicinity of 12 K, the measured resistance

was constant within experimental resolution ( $\sim 1 \times 10^{-4}$ ); at higher temperatures, the resistance exhibited a slow, *irreversible* decrease. Annealing the films at  $\sim 16$  K made the normal-state resistance more nearly constant, but even after annealing a small *reversible* decrease still remained (Fig. 2). Korn *et al.*<sup>21</sup> have made a detailed study of the temperature dependence of the normal-state resistance in amorphous superconductors. Their measurements on amorphous Bi + 25 at. % Ag films are in agreement with the present measurements on amorphous Bi + 3 at. % Tl. Furthermore, their results show that the normal-state resistance varies *linearly* with temperature. This is in contrast with measurements made on thin polycrystalline Al films where the temperature dependence appears to be exponential.<sup>22</sup>

All films used in this study were annealed before any measurements were made. Several of the films were made with the composition Bi + 10 at. % Tl; these films could be annealed at  $\sim 100$  K without crystallizing. After annealing, the normal-state resistance  $R_n(T)$  could be represented by

$$R_n(T) = R_n(0)(1 - aT), \quad (8)$$

where  $a \approx 10^{-4} \text{K}^{-1}$ . Thus, over the range in which the data could be analyzed to determine the fluctuation conductivity ( $6 \text{ K} \leq T \leq 9 \text{ K}$ ) the *change* in  $R_n(T)$  was negligible.

The primary difficulty in determining the residual resistance  $R_0$  is due to the finite magnitude of the fluctuation conductivity at the highest temperatures at which measurements could be made without crystallizing the Bi + 3 at. % Tl films used in this study; thus,  $R_{\max} = R(12 \text{ K})$  was less than  $R_n(12 \text{ K})$ . The fluctuation conductivity can be estimated from Eq. (5) if  $\eta$  is known. Using  $\eta \approx 90 \text{ \AA}$

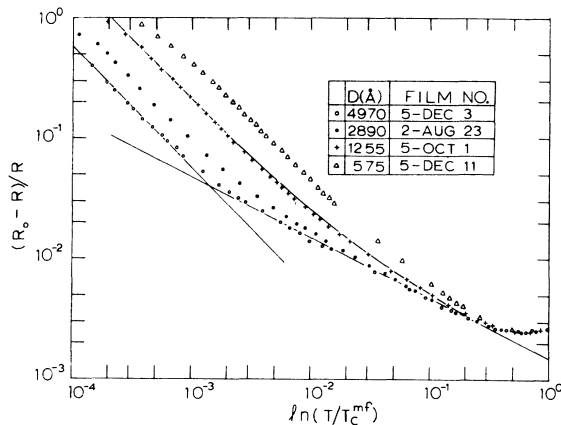


FIG. 4. Temperature dependence of the fluctuation conductivity. Data are plotted for four films;  $R_{0\Box}$ ,  $\tau_0/R_{0\Box}$ ,  $T_c^{mf}$  and  $\eta$  are given in Table II. The curved solid line was calculated from the AL theory with finite thickness corrections [Eq. (1)]. The two straight lines give the limiting two- ( $\sigma' \propto \tau^{-1}$ ) and three-dimensional ( $\sigma' \propto \tau^{-1/2}$ ) forms of the AL theory. Error bars are the size of the plotted points.

as determined from critical-field measurements on the films used in this study,<sup>19</sup> one can estimate that  $\sigma'_{AL}(12\text{ K}) \approx 2 \times 10^{-3} \sigma_n$ ; thus,

$$R_n(12\text{ K}) = R(12\text{ K})(1 + \sigma'/\sigma_n) \approx 1.0020 R_{\max}.$$

This is the basis for determining  $R_0$  from the relation  $R_0 = 1.0025 R_{\max}$ . (The additional factor of  $5 \times 10^{-4}$  accounts for the increase in  $R_n$ , when going from 12 down to  $\sim 7$  K.) The fact that this same procedure for determining  $R_0$  gave consistent results for *all* the films studied is a strong reason for believing it to be valid.

## VI. DISCUSSION

### A. Temperature dependence of the fluctuation conductivity

The most noteworthy aspect of the present results is the definitive observation of the two- to three-dimensional transition predicted by the AL theory. Except perhaps for the thinnest films, each film exhibited a two-dimensional region ( $\sigma' \propto \tau^{-1}$ ) and, at higher temperatures, a three-dimensional region ( $\sigma' \propto \tau^{-1/2}$ ).

The results agree closely with Eq. (1) for  $0.0003 \leq \ln T/T_c^{mf} \leq 0.3$ . Deviations near  $T_c^{mf}$  are believed due to the expected breakdown of the AL theory in the critical fluctuation region.<sup>23</sup> Deviations at  $T \geq 2T_c$  are due to the previously discussed temperature dependence of the normal-state resistance. Small systematic differences between experiment and theory were observed in the vicinity of the two- to three-dimensional transition. These deviations most likely result from the idealized boundary conditions assumed in the derivation of Eq. (1).<sup>15</sup>

The experimentally determined temperature dependence in the three-dimensional region is dependent on the value used for  $R_0$ . For  $R_0 = 1.0025 R_{\max}$ ,  $\sigma'_{3D} \propto \tau^{-n}$  where  $n = 0.50 \pm 0.01$ ; increasing (decreasing)  $R_0$  by  $1 \times 10^{-3}$  changes  $n$  to 0.44 (0.56). Larger changes in  $R_0$  would not be consistent with the measurements of  $R_n(T)$  discussed in Sec. V.

### B. Experimental values of $\tau_0/R_{0\Box}$

The AL theory predicts a universal value of  $1.52 \times 10^{-5} \Omega^{-1}$  for  $\tau_0/R_{0\Box}$ . The present results are 5% to 17% higher than this value. Figure 5 shows the experimental values for a number of amorphous Bi films. The largest error in the determination of  $\tau_0/R_{0\Box}$  as well as the greatest enhancement over the theoretical value occurs for thick films (small  $R_{0\Box}$ ). This is, to some extent, an artifact of the graphical procedure used to determine  $\tau_0$ . Even discounting the highest values of  $\tau_0/R_{0\Box}$ , the experimental values average about

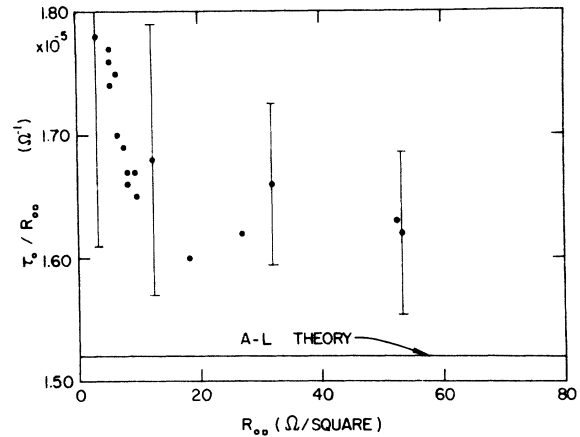


FIG. 5. Comparison of measured and theoretical values of the AL parameter  $\tau_0/R_{0\Box}$  for amorphous Bi films.

10% greater than the AL value. The enhancement of  $\tau_0$  could result from residual strains or inhomogeneities in the films. However, the uniformity and consistent characteristics of amorphous Bi films argue against this possibility. Another possibility is the strong coupling nature of amorphous Bi. Estimates of the effect of strong coupling on the fluctuations, however, predict a decrease rather than an enhancement of  $\tau_0$ .<sup>24,25</sup> A final possibility has been suggested by Fulde and Maki<sup>24</sup> who predict that pair-breaking effects should enhance the value of  $\tau_0$ . The presence of pair breaking in amorphous Bi is suggested by the apparent lack of a contribution from the Maki-Thompson term.<sup>4,5</sup>

### C. Experimental values of $\eta$

Eliminating the results from the thinnest films, the  $\eta$  values determined by fitting the data to Eq. (1) were remarkably consistent, averaging approximately 100 Å. By way of comparison, Lejeune and Naugle<sup>26</sup> have determined  $\eta$  from the proximity effect between amorphous Bi and Fe, finding a value of  $95 \pm 6$  Å. Bergman<sup>27</sup> has calculated a value of 70 Å from critical-field measurements on amorphous  $\text{Bi}_{0.85}\text{Tl}_{0.15}$ . The author has found a value of 90 Å from critical-field measurements on Bi + 3 at. % Tl.<sup>19</sup> A value of  $72 \pm 9$  Å was determined by Glover<sup>14</sup> from measurements of the fluctuation conductivity in the presence of a perpendicular magnetic field.

### D. Comparison with previous measurements on amorphous Bi

The present results are best compared with Glover and Naugle's<sup>14</sup> measurements on two amorphous Bi films for which they reported thicknesses of 2750 and 3200 Å, with  $\tau_0/R_{0\Box}$  equal to (1.46

$\pm 0.11) \times 10^{-5} \Omega^{-1}$  and  $(1.43 \pm 0.11) \times 10^{-5} \Omega^{-1}$ , respectively, and  $\eta$  equal to  $86 \pm 22 \text{ \AA}$  and  $80 \pm 20 \text{ \AA}$ , respectively. Using these values they found quantitative agreement with Eq. (1). In analyzing their data Glover and Naugle adjusted  $R_{00}$ ,  $\tau_0$ , and  $\eta$  so as to optimize the fit to Eq. (1) over the entire measurement range. This procedure differs substantially from the present method; however, the results obtained by the two analysis procedures agree within the combined errors.

#### E. Contribution from the Maki-Thompson fluctuation mechanism

It has been assumed in this paper that the Maki-Thompson fluctuation mechanism<sup>4,5</sup> does not contribute to the fluctuation conductivity in amorphous Bi. The reasons behind this assumption are discussed in detail elsewhere.<sup>19</sup> Briefly, they are as follows: (i) To fit the data requires  $\delta > 1$  for the thinner films, where  $\delta$  is Thompson's pair-breaking parameter.<sup>5</sup> (ii) Fitting the data to the Maki-Thompson theory results in  $\eta \sim 300 \text{ \AA}$ , which is far too large when compared with other determinations of  $\eta$ , all of which give values below  $\sim 100 \text{ \AA}$ .<sup>14,19,26,27</sup> (iii) Recent theoretical results have shown that the electron-phonon interaction rate is greatly enhanced in amorphous metals, leading to a suppres-

sion of the Maki-Thompson fluctuation mechanism.<sup>28</sup>

#### VII. SUMMARY

Measurements of the superconducting fluctuation contribution to the conductivity of amorphous Bi films gave results in agreement with the Aslamazov-Larkin (AL) theory<sup>3</sup> for temperatures in the range  $0.0003 \lesssim \ln T/T_c \lesssim 0.30$ . Films in the thickness range  $500 \lesssim D \lesssim 5000 \text{ \AA}$  agreed with the AL prediction for the temperature and thickness dependence of the fluctuation conductivity; all but the thinnest films clearly showed the change from two-dimensional to three-dimensional behavior predicted by the AL theory.

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