

FLUCTUATION EFFECTS IN THE ac CONDUCTIVITY OF THIN LEAD FILMS ABOVE THE  
SUPERCONDUCTING TRANSITION TEMPERATURE\*

S. L. (A.) Lehoczky†

McDonnell Research Laboratories, McDonnell Douglas Corporation, St. Louis, Missouri 63166

and

C. V. Briscoe

University of North Carolina at Chapel Hill, Chapel Hill, North Carolina 27514

(Received 11 August 1969)

The transmission of microwaves through thin ( $\sim 100\text{-\AA}$ ) Pb films indicates considerable excess conductivity at temperatures above the superconducting transition temperature. Both the frequency and temperature dependences of this conductivity are in agreement with a calculation of Schmidt which takes account of the fluctuations of the order parameter for a film of thickness less than the Ginzburg-Landau coherence length. This agreement between experiment and theory supports Schmidt's choice of time-dependent Ginzburg-Landau equations.

Recently considerable attention has been given to the study of superconductors above the transition temperature  $T_c$  for situations in which the effective electron mean free path is small compared with the Ginzburg-Landau (GL) coherence length.<sup>1</sup> The excess ac conductivity above  $T_c$  due to fluctuations in the order parameter for small mean-free-path superconductors has been investigated theoretically by Schmidt.<sup>2</sup> D'Aiello and Freedman<sup>3</sup> have reported microwave transmission data at 20 GHz on granular Al films which are not in agreement with Schmidt's theory. In this paper we report results of microwave transmission measurements on thin ( $\sim 100\text{-\AA}$ ) Pb films which are in very good accord with the results of Schmidt.

Previous experimental studies<sup>4</sup> have demonstrated an excess dc conductivity above  $T_c$  for a variety of small mean-free-path films which have been amorphous, dirty, granular, or thin (or combinations of these). In the majority of these studies the resistance varies as

$$R(T) = R_N (1 + \tau_0/\tau)^{-1}, \quad (1)$$

where  $R_N$  is the normal-state resistance,  $\tau = (T - T_c)/T_c$ , and  $\tau_0$  is a constant. Aslamazov and Larkin<sup>5</sup> first derived Eq. (1) for disordered two-dimensional metallic systems (a superconducting film with thickness less than the GL coherence length should satisfy this condition reasonably well). According to these authors  $\tau_0/R_N$  should be a universal constant equal to  $0.152 \times 10^{-4} \Omega^{-1}$  regardless of the origin of the specimens. More recent theoretical work<sup>6,7</sup> in this area is basically in agreement with that of Aslamazov and Larkin although there are some systems in which discrepancies between theory and

experiment have been reported.<sup>7</sup>

For the ac case Schmidt,<sup>2</sup> using the time-dependent GL equations given by Abrahams and Tsuneto,<sup>8</sup> obtains (for a film of thickness  $d$  much less than the GL coherence length), for the excess conductivity parallel to the film,

$$\sigma_{\parallel}(\omega, T) = \frac{e^2}{16\hbar d \tau} \left[ \frac{2}{\tilde{\omega}} - \frac{2}{\tilde{\omega}} \tan^{-1} \frac{1}{\tilde{\omega}} - \frac{1}{\tilde{\omega}^2} \ln |1 + \tilde{\omega}^2| \right], \quad (2)$$

where  $\tilde{\omega} = \pi \hbar \omega / 16 k_B T_c \tau$ .

Our measurements at 23.9, 37.2, and 69.6 GHz were made on films prepared from 99.999% purity Pb. The evaporation and subsequent measurements were performed within a microwave cryostat.<sup>9</sup> The Pb was evaporated from a point-source Mo oven and condensed onto  $Z$ -cut, optically polished, quartz plates at 77°K. The flux of the evaporating ions was highly collimated to assure uniformity along the large dimension of the film. In order to obtain stable thin films a slow deposition rate ( $1 \text{ \AA}/\text{min}$ ) was used. Hence, in spite of the rather good vacuum ( $< 10^{-6}$  Torr) the films were probably quite dirty because of absorbed gas molecules. Following the deposition of a given film, it was thermally annealed by allowing the sample block to warm to room temperature. Both dc and microwave measurements were then made simultaneously on the films without breaking the vacuum. Generally, the agreement between the resistance obtained from dc measurements and that computed from the normal-state transmission coefficients was better than 10%, thereby indicating a considerable uniformity in the resistivity and thickness

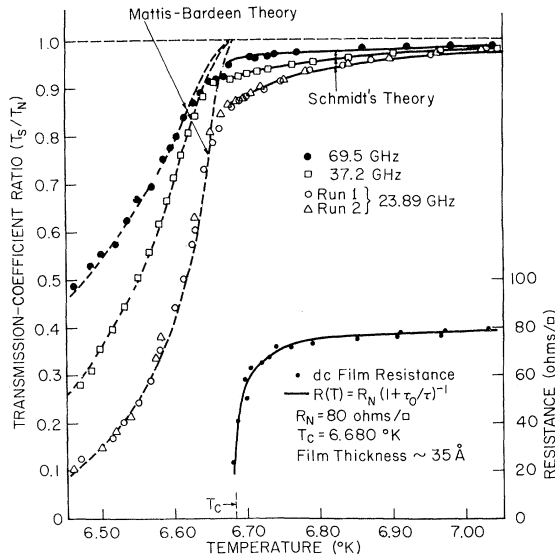


FIG. 1. Microwave transmission-coefficient ratios for a thin Pb film at three frequencies near the superconducting transition temperature. The curve for the dc resistance  $R(T)$  was calculated using the  $R_N$  and  $T_c$  given in the legend and assuming  $\tau_0/R_N = 0.152 \times 10^{-4} \Omega^{-1}$ .

over a major portion of the films. The thicknesses of the films as estimated from the residual resistance values ranged from 30 to 150 Å. The effective mean free paths  $l_{eff}$  computed in similar fashion varied from 16 to 31 Å. Apparently, the films were well within the criteria for

dirty superconductors (i.e.,  $l_{eff} \lesssim \xi_0$ , where  $\xi_0$  is the coherence length in the pure material).

In Fig. 1 the microwave transmission-coefficient ratio  $T_S/T_N$  is shown as a function of temperature for a film of 35 Å effective thickness. Because of the possibility of induced currents in the films exceeding critical values near  $T_c$ , the microwave power was steadily lowered until the experimental curves showed no dependence upon power level. The microwave power then was further reduced by a factor of 100 to obtain the data presented here. As reported previously,<sup>10</sup> the results below  $T_c$  are in good agreement at all three frequencies with the ac conductivity equations derived by Mattis and Bardeen<sup>11</sup> from the BCS theory if  $4.5 \pm 0.2$  is used for  $2\Delta_0/kT_c$ . From infrared-transmission measurements on Pb thin films similar to ours, Palmer and Tinkham<sup>12</sup> obtain  $2\Delta_0/kT_c = 4.5 \pm 0.1$ .

As can be seen in Fig. 1 the data near and above  $T_c$  deviate considerably from those expected on the basis of the Mattis and Bardeen theory. In this temperature region we analyzed the data considering the effects on  $T_S/T_N$  of the additional contributions to the dynamic conductivity proposed by Schmidt. In this case, the total conductivity is  $\sigma(\omega, T) = \sigma_{||}(\omega, T) + \sigma_N$ , where  $\sigma_{||}(\omega, T)$  is given by Eq. (2) and  $\sigma_N$  is the residual value of the conductivity in the normal state. Thus,

$$\frac{\sigma(\omega, T)}{\sigma_N} = 1 + \frac{e^2 R_N}{16\hbar\tau} \left[ \frac{\pi}{\omega} - \frac{2}{\omega} \tan^{-1} \frac{1}{\omega} - \frac{1}{\omega^2} \ln |1 + \tilde{\omega}^2| \right].$$

The transmission-coefficient ratio is given by<sup>9</sup>

$$T_S/T_N = \{n^2[2 + Z_g/R_N]^2 \cos^2 kl + [n^2 + 1 + Z_g/R_N]^2 \sin^2 kl\} \{n^2[(2 + Z_g\sigma_1/R_N\sigma_N)^2 + (Z_g\sigma_2/R_N\sigma_N)^2] \cos^2 kl + [(n^2 + 1 + Z_g\sigma_1/R_N\sigma_N)^2 + (Z_g\sigma_2/R_N\sigma_N)^2] \sin^2 kl - n(n^2 - 1)(Z_g\sigma_2/R_N\sigma_N) \sin 2kl\}^{-1}, \quad (4)$$

where  $Z_g$  is the waveguide impedance,  $k$  is the propagation constant in the quartz substrate,  $l$  is the substrate thickness,  $N$  is the index of refraction of quartz, and  $\sigma_1$  and  $\sigma_2$  are the real and imaginary parts of the superconducting complex conductivity, respectively. The solid lines above  $T_c$  in Fig. 1 (labeled Schmidt's theory) were calculated using  $\sigma(\omega, T)/\sigma_N$  for  $\sigma_1/\sigma_N$  and  $\sigma_2/\sigma_N = 0$  in Eq. (4) above. The agreement of experiment and theory with respect to both temperature and frequency is quite good. It is worth noting that runs 1 and 2 at 23.89 GHz occurred two weeks apart in time.

To allow a more critical comparison between the theory and the experimental results, the temperature and frequency dependence of  $\sigma(\omega, T)/$

$\sigma_N$  above  $T_c$ , computed from the inversion of Eq. (4), is shown in Fig. 2. The experimental points in Fig. 2 have been multiplied by small normalization factors; for 69.6 GHz the factor was 1.005, for 23.89 GHz it was 1.017 (run 1) and 1.016 (run 2), and for 37.2 GHz it was 1.0185. These factors are of no consequence on the scale of Fig. 1. These normalization factors essentially are corrections to the measured values of  $T_N$ . When these measurements were made we were interested primarily in the temperature range below  $T_c$ . Consequently, data were not taken at temperatures far enough above  $T_c$  to obtain precise values of  $T_N$ . Even so, in this type of

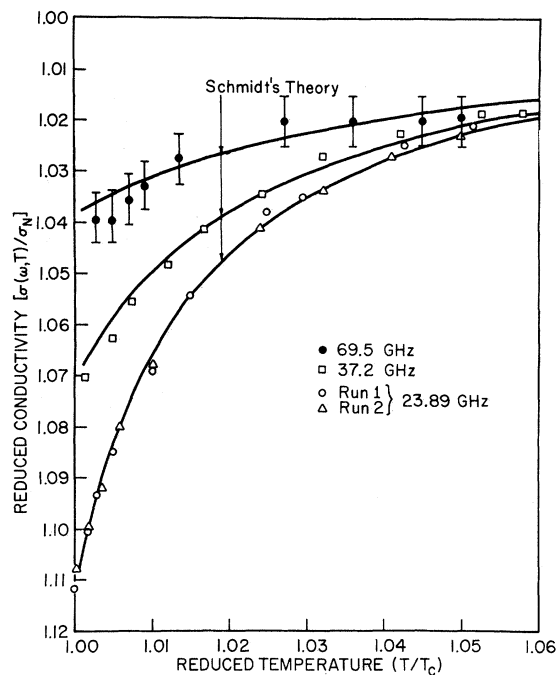


FIG. 2. Reduced ac conductivities calculated from the data shown in Fig. 1. Error bars for the two lower frequencies are about twice the size of the data points.

experiment the limitations imposed by the long-range stability of the microwave measuring equipment would make it extremely difficult to eliminate discrepancies of such small magnitude.

In conclusion, the good agreement between the ac-conductivity theory of Schmidt and the experimental data as a function of both frequency and temperature indicates that the basic assumptions (in particular the form of the time-dependent Ginzburg-Landau equations) and procedures of the theory are satisfactory for thin, dirty Pb films.

We are grateful to H. Schmidt for sending us a copy of his paper before publication.

\*Research supported by the University of North Carolina Materials Research Center under Contract No. SD-100 from the Advanced Research Projects Agency.

†The McDonnell Research Laboratories contribution to this paper was performed under the McDonnell Douglas Independent Research and Development Program.

<sup>1</sup>See "Search and Discovery," *Phys. Today* **22**, No. 5, 57 (1969).

<sup>2</sup>H. Schmidt, *Z. Physik* **216**, 336 (1968).

<sup>3</sup>R. V. D'Aiello and S. J. Freedman, *Phys. Rev. Letters* **22**, 515 (1969).

<sup>4</sup>R. E. Glover, *Phys. Letters* **25A**, 542 (1967); M. Strongin, O. F. Kammerer, J. E. Crow, R. S. Thompson, and H. L. Fine, *Phys. Rev. Letters* **20**, 922 (1968); R. O. Smith, B. Serin, and E. Abrahams, *Phys. Letters* **28A**, 224 (1968).

<sup>5</sup>L. G. Aslamazov and A. I. Larkin, *Fiz. Tverd. Tela* **10**, 1104 (1968) [translation: *Soviet Phys.—Solid State* **10**, 875 (1968)], and *Phys. Letters* **26A**, 238 (1968).

<sup>6</sup>E. Abrahams and J. W. F. Woo, *Phys. Letters* **27A**, 117 (1968); P. Fulde and K. Maki, in *Proceedings of the Eleventh International Conference on Low Temperature Physics, St. Andrews, Scotland, 1968*, edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (St. Andrews University, St. Andrews, Scotland, 1968); L. P. Kadanoff and G. Laramore, *Phys. Rev.* **175**, 579 (1968); H. Schmidt, *Phys. Letters* **27A**, 658 (1968); R. A. Ferrell and H. Schmidt, *Phys. Letters* **25A**, 544 (1967); J. P. Hurault, *Phys. Rev.* **179**, 494 (1969).

<sup>7</sup>See L. R. Testardi, W. A. Reed, P. C. Hohenberg, W. H. Haemmerle, and G. F. Brennert, *Phys. Rev.* **181**, 800 (1969), for a critical discussion of data analysis and comparison with theory for most of the work in this area including their own.

<sup>8</sup>E. Abrahams and T. Tsuneto, *Phys. Rev.* **152**, 416 (1966).

<sup>9</sup>N. M. Rugheimer, A. Lehoczy, and C. V. Briscoe, *Phys. Rev.* **154**, 414 (1967).

<sup>10</sup>A. Lehoczy and C. V. Briscoe, *Bull. Am. Phys. Soc.* **13**, 729 (1968).

<sup>11</sup>D. C. Mattis and J. Bardeen, *Phys. Rev.* **111**, 412 (1958).

<sup>12</sup>L. H. Palmer and M. Tinkham, *Phys. Rev.* **165**, 588 (1968).

## EFFECT OF ELECTRON-LO-PHONON COUPLING ON RAMAN SCATTERING IN CADMIUM SULFIDE

G. P. Vella-Coleiro

Bell Telephone Laboratories, Murray Hill, New Jersey 07974

(Received 22 July 1969)

Enhanced Raman scattering by LO phonons in CdS has been observed when the energy difference between Landau levels in the conduction band is equal to the LO phonon energy at the zone center.

Oscillations in the conductivity due to coupling between electrons and longitudinal optical phonons have recently been observed in several

semiconductors.<sup>1-5</sup> In the presence of a transverse magnetic field, minima in the conductivity occur when the energy difference between two