

Microwave Transmission- and Reflection-Coefficient Ratios of Thin Superconducting Films*

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 (Received 20 June 1966)

Measurements of the transmission and reflection of 12-, 8-, and 4-mm microwaves through thin ($\sim 50 \text{ \AA}$) superconducting films of tin and indium were made as a function of temperature below the superconducting transition temperature. A vacuum microwave cryostat with the waveguide passing straight through the system was used to make the measurements. A movable sample block made it possible to condense and anneal the films and make microwave measurements without removing the films from the vacuum. Superconducting complex electrical conductivities as formulated by Mattis and Bardeen were used to compute transmission- and reflection-coefficient ratios for comparison with experimental values. In the majority of cases the agreement between experiment and theory is quite good. In some cases the assumption of a superconducting energy gap of $2\Delta(0) = 2.82kT_c$, instead of the predicted $2\Delta(0) = 3.52kT_c$, improved the fit between experiment and theory. The temperature at which the microwave transmission and reflection differ measurably from their values in the normally conducting states can be used as a definition of the critical temperature. Transition temperatures measured in this manner are in close agreement with the usual dc definition.

I. INTRODUCTION

IN these experiments thin films of tin and indium, which were condensed onto quartz substrates, were placed directly across a waveguide. Measurements of the microwave transmission and reflection coefficients of these films in both the normal and superconducting states furnish direct evidence of the changes in electrical conductivity that occur as a film is cooled below its transition temperature. Idealizing the actual situation to that of a film of thickness d (\ll wavelength or skin depth) deposited on a substrate of thickness l and index of refraction n , the transmission and reflection coefficients for the case of normal incidence can be calculated exactly. It is convenient to introduce, following Glover and Tinkham,¹ a complex conductivity $\sigma = \sigma_1 - i\sigma_2$ for the superconducting state. For the case in which the radiation is incident on the film before the substrate, the transmission and reflection coefficients in the superconducting state are

$$T_S = 4n^2 \{ n^2 [(2 + Z_\theta \sigma_1 d)^2 + (Z_\theta \sigma_2 d)^2] \cos^2 kl + [(n^2 + 1 + Z_\theta \sigma_1 d)^2 + (Z_\theta \sigma_2 d)^2] \sin^2 kl + n(n^2 - 1) Z_\theta \sigma_2 d \sin 2kl \}^{-1} \dots \quad (1)$$

and

$$R_S = \{ n^2 [(Z_\theta \sigma_1 d)^2 + (Z_\theta \sigma_2 d)^2] \cos^2 kl + [(n^2 - 1 + Z_\theta \sigma_1 d)^2 + (Z_\theta \sigma_2 d)^2] \sin^2 kl + n(n^2 - 1) Z_\theta \sigma_2 d \sin 2kl \} / \{ n^2 [(2 + Z_\theta \sigma_1 d)^2 + (Z_\theta \sigma_2 d)^2] \cos^2 kl + [(n^2 + 1 + Z_\theta \sigma_1 d)^2 + (Z_\theta \sigma_2 d)^2] \sin^2 kl + n(n^2 - 1) Z_\theta \sigma_2 d \sin 2kl \} \dots, \quad (2)$$

where Z_θ is the waveguide impedance and k is the propagation constant in the quartz substrate. The transmission and reflection coefficients in the normal state T_N

and R_N can be found by setting $\sigma_1 = \sigma_N$ and $\sigma_2 = 0$ in the above equations. These equations can be inverted to give the conductivities as functions of the transmission and reflection coefficients.

$$Z_\theta \sigma_1 d = n^2 (1 - R_S - T_S) / [T_S (n^2 \cos^2 kl + \sin^2 kl)] \dots \quad (3)$$

$$Z_\theta \sigma_2 d = \{ n(1 - n^2) \sin kl \cos kl + [(4n^2 / T_S) \times (n^2 \cos^2 kl + \sin^2 kl) - \{ n^2 \cos^2 kl + \sin^2 kl + (n^2 / T_S)(1 - R_S) \}^2]^{1/2} \} / (n^2 \cos^2 kl + \sin^2 kl) \dots \quad (4)$$

Mattis and Bardeen² have derived formulas using the BCS³ theory which enable one to calculate σ_1 / σ_N and σ_2 / σ_N . These conductivities are functions of the frequency of the incident electromagnetic wave, the transition temperature, the absolute temperature, and the superconducting energy gap. In Fig. 1 σ_1 / σ_N and σ_2 / σ_N are shown for four microwave frequencies which span the frequency range used in the experiments reported in this paper.

The following constants were used in a typical calculation of T_S and R_S : microwave frequency = 23.8 GHz, $Z_\theta = 473.4 \Omega$, $n = 2.07$, $l = 0.038 \text{ cm}$, $d = 28 \text{ \AA}$, $k = 8.22 \text{ cm}^{-1}$, and $\sigma_N = 4.76 \times 10^4 \Omega^{-1} \text{ cm}^{-1}$. If the conductivities corresponding to $\hbar\omega = 0.25kT_c$ shown in Fig. 1 are used the resulting T_S / T_N and R_S / R_N curves were almost identical to the theoretical curves shown in Fig. 7(a).

Measurements of the ratios of the transmission-coefficients in the superconducting state to that in the normal state were made. Similar measurements were made on the reflection coefficients. These ratios are referred to hereafter as the transmission- and reflection-coefficient ratios, T_S / T_N and R_S / R_N , respectively. In a given experiment, T_S / T_N and R_S / R_N were measured as functions of temperature at constant microwave frequency. The results are compared with the theoretical

* Work partly supported by the Advanced Research Projects Agency.

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¹ R. E. Glover, III and M. Tinkham, Phys. Rev. **108**, 243 (1957).

² D. C. Mattis and J. Bardeen, Phys. Rev. **111**, 412 (1958).

³ J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

values of the ratios calculated using the conductivities of Mattis and Bardeen² in Eqs. (1) and (2) above.

A preliminary account of this work was presented at the Ninth International Conference on Low-Temperature Physics.⁴

II. EXPERIMENTAL APPARATUS AND PROCEDURE

Microwave Equipment

In these experiments measurements were made in the frequency ranges 22–26, 28–35, and 67–73 GHz. The microwaves were generated by 12, 8, and 4 mm reflex klystrons. The microwave apparatus was similar in each case. A reflectometer arrangement was used to measure the incident and reflected power. The transmitted power was measured in a similar manner. The output power of the 4-mm klystron was about 150 mW. The 12- and 8-mm klystrons were rated at 40 mW. Sufficient attenuation was placed in the waveguide so that the power incident on the film was less than 1 mW in most cases. The frequencies were monitored by means of cavity wave meters.

Cryostat

The vacuum microwave cryostat used in this experiment was versatile enough that the film could be made, annealed, and moved into place directly across the waveguide without breaking the vacuum. The waveguide passed “straight through” the cryostat. (See Fig. 2).

Just before the waveguide entered the cryostat a transition piece from the rectangular waveguide to the circular waveguide was placed in the line. At this junction two polyethylene windows (0.0025 in. thick)

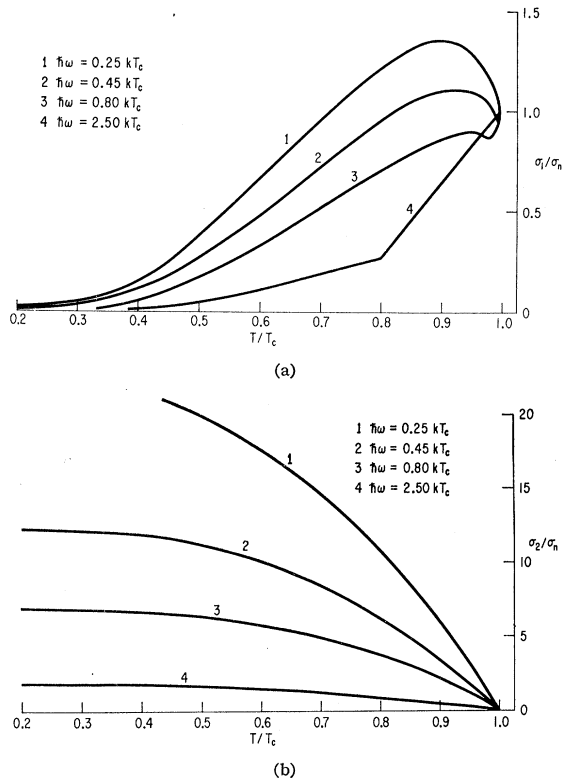
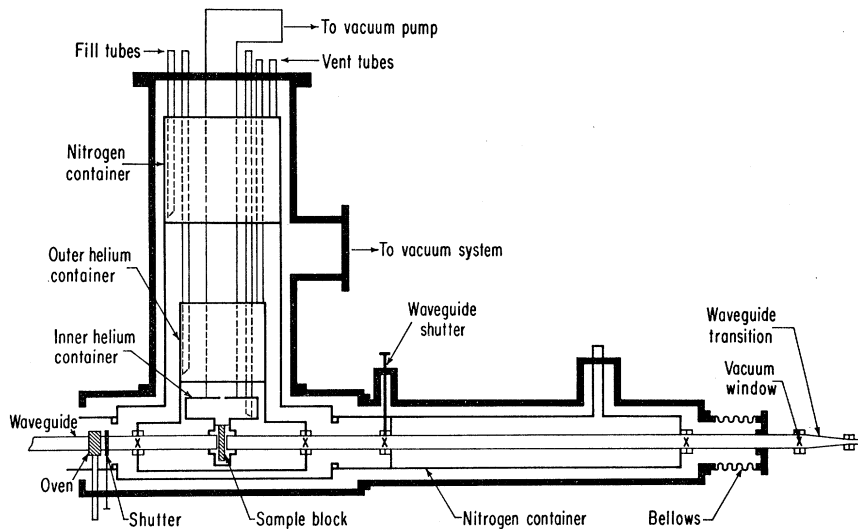


FIG. 1. Complex conductivities calculated from the formulas derived by Mattis and Bardeen (Ref. 2).

separated by approximately one-quarter wavelength served as a vacuum seal. Inside the cryostat, $\frac{1}{2}$ -in. o.d. (0.010 in. wall) stainless-steel tubing was used as waveguide. After leaving the cryostat, another transition piece returned the guide from circular to rectangular.

FIG. 2. Schematic drawing of the microwave cryostat in which the films could be made, annealed, and moved into position directly across the waveguide without breaking the vacuum. The left extension arm, identical to the one illustrated, is not shown in the drawing.



⁴ N. M. Rugheimer, C. V. Briscoe, A. Lehoczky, and R. E. Glover, III, *Proceedings of the Ninth International Conference on Low Temperature Physics* (Plenum Press, New York, 1965) p. 381.

Two heat shields, one at liquid-nitrogen temperature and the other at liquid-helium temperature, completely surrounded the sample block except for the entrance and exit ports of the waveguide. To minimize the radiation from these two ports, two extension arms were added to the microwave line so that the waveguide was maintained at liquid-nitrogen temperature from 10 to 24 in. on either side of the sample block. Several thin polyethylene windows were placed in the circular waveguide to further reduce the heat exchange via the waveguide between the sample and the outside surroundings. These windows also helped to minimize the effect of resonances in the voltage standing-wave ratio of the line as a function of frequency. The first 4 in. of waveguide on either side of the sample block was cooled by the helium radiation shield.

Temperatures from 4.2° to 1.1°K were maintained by adjusting the vapor pressure above the liquid helium in the inner reservoir. The vapor pressure was measured by an ordinary U-tube manometer with the open end connected to the helium reservoir. The temperature was also measured using a carbon resistance thermometer imbedded in the sample block.

The substrates were *z*-cut natural quartz plates 1.24 in. × 2.00 in. × 0.015 in. with the optical axis perpendicular to the surface on which the film was condensed. (See Fig. 3.) The large surfaces of the quartz were optically polished, and the variation in these surfaces did not exceed ± 0.0005 in.

The films were vacuum condensed onto the substrate through a mask. The mask was positioned so that the film covered approximately one-half of the substrate. (See Fig. 3.) The sample block could be moved by a gear drive mechanism into three different positions, so that the microwaves would pass through a hole in the sample block, through an uncovered section of the quartz, or through both the film and the quartz.

Procedure

After chemical cleaning, the substrate was placed in a small auxiliary vacuum chamber where gold electrodes were condensed onto it. The substrate was then transferred to the sample block in the cryostat and the apparatus was assembled. After the system had been evacuated to $<10^{-5}$ mm of Hg for more than 12 h, the substrate was cooled to either liquid-nitrogen or liquid-

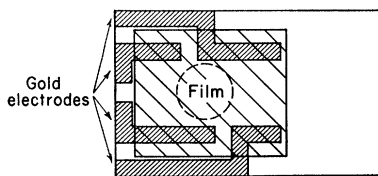


FIG. 3. Schematic drawing of the quartz substrate showing the positions of the gold electrodes and the specimen film. The area of the film outlined by the dashed circle was within the $\frac{1}{2}$ -in. circular waveguide.

helium temperature, and a film was condensed onto it. The film was annealed by allowing the cryostat to warm up to room temperature. After annealing, the cryostat was cooled to liquid-helium temperature, and the transmission- and reflection-coefficient ratios were measured at several constant frequencies in the temperature range 4.2°K to 1.1°K.

III. FILM PARAMETERS

The tin films were prepared by evaporating high purity (0.99999) tin from a tungsten filament and condensing it onto a quartz substrate. The indium films were prepared in a similar manner with the material purity being 0.99999 for indium films 3 and 4 and 0.9997 for indium films 1 and 2. With the exception of tin film 6, all films were electrically continuous.

Tin film 8 and indium film 3, even though electrically continuous, had very large resistances and negative temperature coefficients of resistance. (See Table I). The negative temperature coefficient of resistance implies a conduction mechanism which is thermally

TABLE I. Film parameters.

Film	dc resistance (Ω /square)			Film thickness d (\AA)	Mean free path, l_{eff} (\AA)	Coherence length, ξ (\AA)	T_c ($^{\circ}\text{K}$)
	300°K	77°K	4.2°K				
Tin 3	111.5	84.05	72.18	35	50	49	3.90
Tin 6							3.73
Tin 7	85.10	51.15	43.51	28	90	87	3.95
Tin 8	34 000	53 000	400 000	Tunneling film			
Indium 1	14.07	7.50	6.19	107	241	228	3.85
Indium 2	61.95	45.35	39.20	42	108	101	3.93
Indium 3	21 <i>M</i>	40 <i>M</i>		Tunneling film			
Indium 4	488	405	339	8.4	82	81	3.98

assisted, e.g., tunneling. These two films were probably made up of many small clumps or islands of metal separated by distances small enough to permit tunneling of electrons from one clump of metal to another. If this model is correct, the current-voltage characteristics of these films should be similar to the current-voltage characteristics of a single superconducting tunnel junction. (See Giaever.⁵) The current-voltage characteristics of tin film 8 are shown in Fig. 4 for various temperatures. The similarity to the current-voltage characteristics of a single superconducting tunnel junction is apparent. The abscissa scale of tens of volts rather than mV implies many tunnel junctions in series (possibly as many as ten thousand). These two films appeared continuous when viewed through an oil immersion microscope and the microwave measurements were similar to those made on other very thin films.

Several parameters which are useful in describing and discussing the properties of thin superconducting films are given in Table I for the films used in this research.

⁵ I. Giaever, Phys. Rev. Letters 5, 147 (1960).

In columns 2, 3, and 4, the dc resistance in ohms per square is given at 300°, 77°, and 4.2°K.

Assuming Matthiessen's rule, the following equation for the thickness d of a film can be derived:

$$d = (l/w)[\partial\rho/\partial T]/(\partial R/\partial T), \quad (5)$$

where l is the length of the film, w is the width of the film, ρ is the resistivity of the material and R is the resistance of the film. The value of $\partial\rho/\partial T$ used for tin was $4.50 \times 10^{-8} \Omega \text{ cm}/^\circ\text{K}$ and the value of $\partial\rho/\partial T$ used for indium was $3.35 \times 10^{-8} \Omega \text{ cm}/^\circ\text{K}$.⁶ The length of the films (the direction of current flow) was 2.02 cm and the width was 2.15 cm. Film thicknesses calculated using the above equation are given in column 5 of Table I. For films of thickness less than 100 Å the results of this type of calculation can be considerably in error⁷; however, in the theoretical equations (1), (2), (3), and (4), the thickness appears only in the products $Z_0\sigma_N d$, $Z_0\sigma_1 d$, $Z_0\sigma_2 d$. Therefore it is not necessary to know the film thickness in order to calculate the ratios σ_1/σ_N and σ_2/σ_N . The power transmitted, however, is proportional to film thickness, and it is convenient to have an estimate of its value.

The ratio of the absolute conductivity σ to the effective electron mean free path l_{eff} is a constant for any metal independent of temperature. From measurements of the ac surface conductivity in the limit of the anomalous skin effect, Chambers⁸ gives for tin a value of $9.5 \times 10^{10} \Omega^{-1} \text{ cm}^{-2}$ for σ/l_{eff} . Toxen *et al.*⁹ give $\sigma/l_{\text{eff}} = 6.25 \times 10^{10} \Omega^{-1} \text{ cm}^{-2}$ for indium as determined from the thickness dependence of the dc electrical resistance of indium films. Using the residual resistance ratio,

$$\rho_r = R(4.2^\circ\text{K})/[R(273^\circ\text{K}) - R(4.2^\circ\text{K})],$$

and the absolute conductivity, $\sigma(273^\circ\text{K})$, we have calculated the electron mean free paths shown in column 6 of Table I. (For the procedure used see, e.g., Chanin *et al.*¹⁰) Values used for $\sigma(273^\circ\text{K})$ were from Ref. 6.

Tinkham¹¹ has suggested that the Pippard coherence length ξ can be calculated from

$$1/\xi = (1/\xi_0) + (1/l_{\text{eff}}) \quad (6)$$

where ξ_0 is the coherence length for bulk material at 0°K and l_{eff} includes effects due to sample dimensions. Faber and Pippard¹² find $\xi_0 = 2100 \text{ Å}$ for tin. Dheer¹³

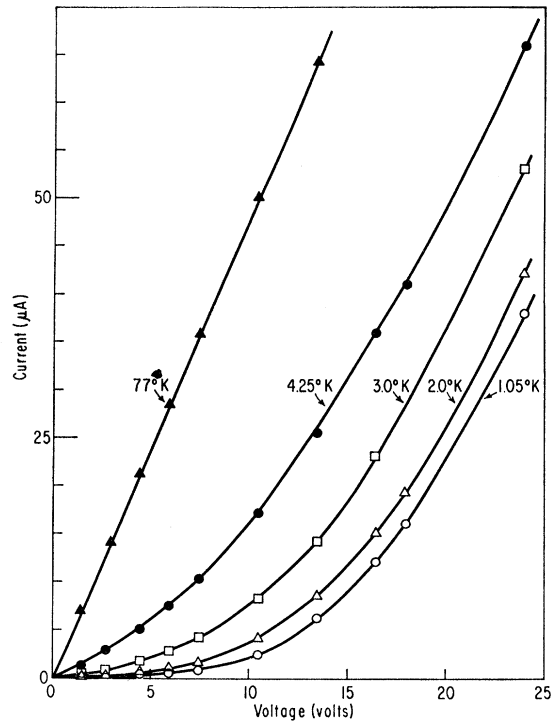


FIG. 4. The current-voltage characteristics of tin film 8 which has a very large resistance and a negative temperature coefficient of resistance. The abscissa scale of volts rather than millivolts implies many superconducting tunnel junctions in series.

gives for indium a value of 4400 Å for ξ_0 . Values for ξ are given in column 7 of Table I.

IV. RESULTS AND DISCUSSION

The results of microwave measurements were similar, although not identical, for different films of the same material. These differences were possibly due to the fact that it is quite difficult to fabricate very thin films with completely identical physical properties. The data, however, taken on any given film were reproducible. The reproducibility of the microwave transmission- and reflection-coefficient ratios was always at least as good as the data shown in Fig. 5. The displacement of T_S/T_N and R_S/R_N for run 2 near T_c was due to a thermal gradient between the sample and the thermometer which resulted when the temperature of the cryostat was allowed to increase at too rapid a rate. The rest of the data presented in this paper were taken as the temperature of the cryostat was varied so slowly that no effects due to thermal gradients were observed.

In fitting the transmission- and reflection-coefficient ratios, calculated using the conductivities of Mattis and Bardeen,² to the experimental data the only free parameter used was the film resistance. The resistance which yielded the best fit between theory and experiment for a particular film was used in all subsequent

⁶ *A Compendium of the Properties of Materials at Low Temperatures (Phase II)*, edited by Richard B. Stewart and Victor J. Johnson (National Bureau of Standards, Cryogenic Engineering Laboratory, Boulder, Colorado, 1961), pp. 321, 341.

⁷ H. Vogel, Ph.D. thesis, the University of North Carolina, Chapel Hill, North Carolina, 1962 (unpublished).

⁸ R. G. Chambers, Proc. Roy. Soc. (London) **A215**, 481 (1952).

⁹ A. M. Toxen, M. J. Burns, and D. J. Quinn, Phys. Rev. **138**, A1145 (1965).

¹⁰ G. Chanin, E. A. Lynton, and B. Serin, Phys. Rev. **114**, 719 (1959).

¹¹ M. Tinkham, Phys. Rev. **110**, 26 (1958).

¹² T. E. Faber and A. B. Pippard, Proc. Roy. Soc. (London) **A231**, 336 (1955).

¹³ P. N. Dheer, Proc. Roy. Soc. (London) **A260**, 333 (1961).

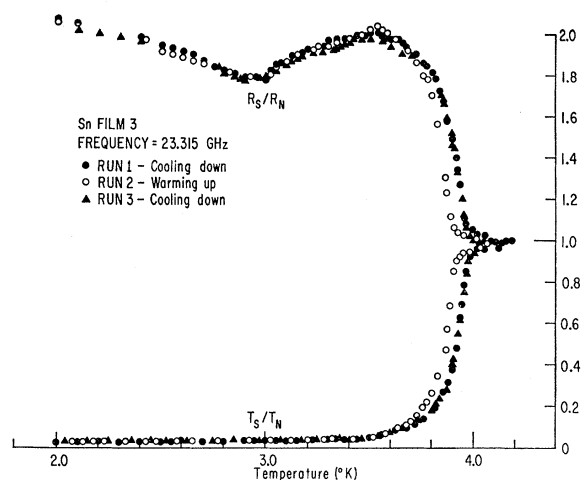


FIG. 5. Temperature dependence of the transmission- and reflection-coefficient ratios for tin film 3 illustrating the reproducibility of the data. The slight displacement of the data for run 2 near T_c was due to a thermal gradient between the sample and the thermometer which resulted when the temperature of the cryostat was allowed to increase at a too rapid rate.

calculations for the same film even though the microwave frequency was different. For all the films, except the tunneling films, this effective resistance used in calculating the transmission- and reflection-coefficient ratios was within a factor of 2.5 of the measured dc resistance.

To facilitate the interpretation of the experimental results it is convenient to classify the energy of the incident electromagnetic radiation depending on whether: (I) $\hbar\omega < 2\Delta(T)$, (II) $\hbar\omega \approx 2\Delta(T)$, or (III) $\hbar\omega > 2\Delta(T)$ for an appreciable range of temperature, where $2\Delta(T)$

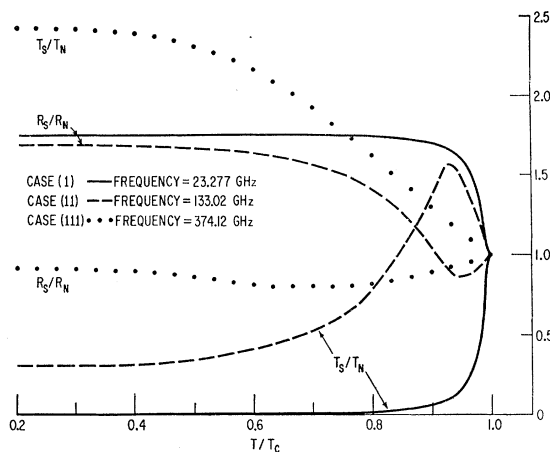


FIG. 6. Theoretical transmission- and reflection-coefficient ratios for a 75- Ω film with a transition temperature of 3.99°K. In case (I) $\hbar\omega = 0.280 kT_c$, in case (II) $\hbar\omega = 1.60 kT_c$, and in case (III) $\hbar\omega = 4.50 kT_c$. At a frequency of 292 GHz, $\hbar\omega = 3.52 T_c$. For cases (I) and (II) T_S/T_N decreases and R_S/R_N increases with decreasing temperature. Note that the temperature dependence of T_S/T_N and R_S/R_N for case (III) is opposite in sign to that of cases (I) and (II).

is the width of the superconducting energy gap at a given temperature. Figure 6 shows transmission- and reflection-coefficient ratios calculated using Eqs. (1) and (2) for these three cases. The range of frequencies available for this work was such that $\hbar\omega < 2\Delta(T)$ for almost all temperatures and frequencies. Thus the majority of the T_S/T_N and R_S/R_N versus temperature curves are basically similar to case (I) shown in Fig. 6.

BCS Behavior

Figures 7 and 8 are examples of T_S/T_N and R_S/R_N for tin and indium films of various thicknesses and at several microwave frequencies. These results are seen to agree well with calculations made using the conductivities of Mattis and Bardeen, assuming a superconducting energy gap of $2\Delta(0) = 3.52 kT_c$. Notice the similarity in all of the data, except that for indium film 2 taken at a frequency of 71.8 GHz, to the theoretical calculation shown in Fig. 6 for the case in which $\hbar\omega$, the microwave energy, is less than $2\Delta(T)$ except for temperatures very near T_c . Figure 7(d) shows the transmission-coefficient ratio for indium film 2 at a microwave frequency of 71.8 GHz which is large enough to excite electrons across the energy gap a measurable distance below T_c . The increase in the transmission coefficient ratio just below T_c is due to the large increase in the density of electron energy states at the gap edge. This is an example of case (II) shown in Fig. 6. Unfortunately no reflection data are available for this film at 71.8 GHz. The discrepancy between the experimental data and the theoretical calculation at low temperature is probably due to a small amount of leakage around the sample film at this relatively high frequency. The higher noise level encountered for the small transmission signal at this frequency makes any correction for leakage somewhat in doubt.

The calculated thickness of tin film 7 was 28 Å which is corroborated by the relatively large value of R_S/R_N (1.76) observed well below T_c . [See Fig. 7(a).] The relatively small value of R_S/R_N (1.25) well below T_c shown in Fig. 7(c) for indium film 2 is consistent with its calculated thickness of 42 Å.

In Fig. 7(b), the results for indium film 1 which had a calculated thickness of 107 Å are shown. The scatter in the data illustrates the difficulty of making good relative measurements of T_S/T_N and R_S/R_N for films thicker than 100 Å. Note the very small change in R_S/R_N in going from the normal to the superconducting state. The transmission-coefficient ratio changes from one in the normal state to essentially zero for temperatures well below T_c . However, the transmission for a relatively thick film (100 Å or greater) is quite small even in the normal state. Hence in dealing with a thick film, one has the problem of a very weak signal (the transmission) which changes considerably, and a strong signal (the reflection) which does not change very much.

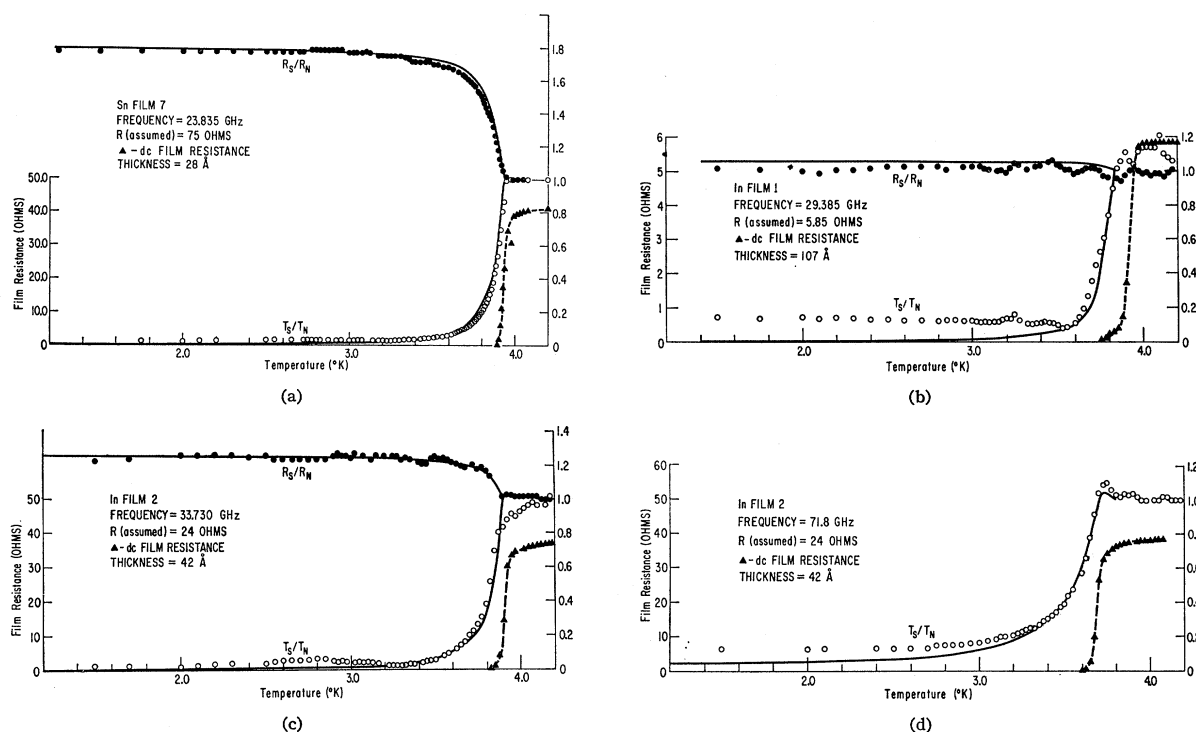


FIG. 7. Temperature dependence of the transmission- and reflection-coefficient ratios for tin film 7 and indium films 1 and 2. The solid lines represent T_S/T_N and R_S/R_N calculated from the BCS theory using the parameters given in the legends. In (d) the increase in T_S/T_N just below the transition temperature is related to the large increase in the BCS density of electron energy states near the gap edge.

This condition leads to considerable scatter in the data as illustrated by the results seen in Fig. 7(b). Note that the scatter in T_S/T_N above T_c is about equal to the discrepancy between T_S/T_N and the theoretical curve at temperatures well below T_c .

The data for indium film 3 are shown in Fig. 8. Even though this was a "tunneling film," the effective ac resistance used in calculating the theoretical curve shown by the solid line was only 70 Ω per square which corresponds to an effective thickness in the range 25 to 50 \AA . Note the large microwave transition temperature, 4.05°K, which is possibly due to strain in the clumps of metal making up the island structure. The dashed line illustrates the effect of increasing the microwave power incident on the film by a factor of 100. The discontinuous changes in the transmission and reflection are the result of the microwave-induced currents exceeding the critical current for the film above 3.675°K. Similar effects have been observed by Gittleman *et al.*¹⁴ at a microwave frequency of 55 GHz for 500 \AA tin films. In their experiment the imaginary part of the microwave impedance was measured as a function of dc current through the films at reduced temperatures of $0.5T_c$ and less. The microwave-induced currents were small

compared to the critical currents. At the critical current, when the tin film became normal, a large change (order of magnitude) in the microwave signal was observed due primarily to a change in the real part of the micro-

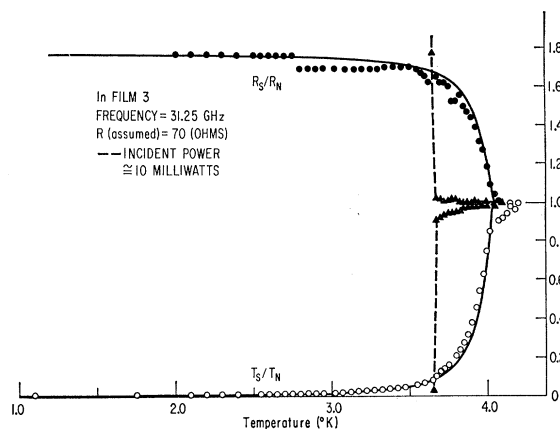


FIG. 8. Temperature dependence of the transmission- and reflection-coefficient ratios for indium film 3. The solid lines represent T_S/T_N and R_S/R_N calculated from the BCS theory using the parameters given in the legend. The triangular points represent data taken at an incident power level of approximately 10 mW, which was at least two orders of magnitude larger than the incident power level used in recording the data represented by the circles. The discontinuous changes in T_S/T_N and R_S/R_N at 3.675°K are due to the microwave-induced current exceeding the critical current for the film above 3.675°K.

¹⁴ J. Gittleman, B. Rosenblum, T. Seidel, and A. Wicklund, *Proceedings of the Eighth International Conference on Low Temperature Physics* (Butterworth and Company, Ltd., London, 1963), p. 336.

wave impedance. In our experiment the dc currents were small compared to the microwave-induced critical currents. This technique should provide a useful means of measuring superconducting critical currents provided one can accurately determine the absolute value of the microwave induced currents in the films. All measurements reported in this paper were made at incident power levels low enough that this discontinuous change in transmission and reflection, if it occurred at all, was so near the transition that it could not be detected.

Reduced Gap

The transmission- and reflection-coefficient ratios measured for tin film 6 did not change as rapidly with temperature as the BCS theory predicts, when calculations were made assuming the width of the gap to be $3.52 kT_c$ at absolute zero. In the BCS theory the energy gap is assumed to be isotropic, i.e., independent of crystallographic direction. For bulk materials and for thin films in general this is usually a reasonable assumption. However, there are references^{15,16} in the literature to tin films which to a certain extent exhibit single-

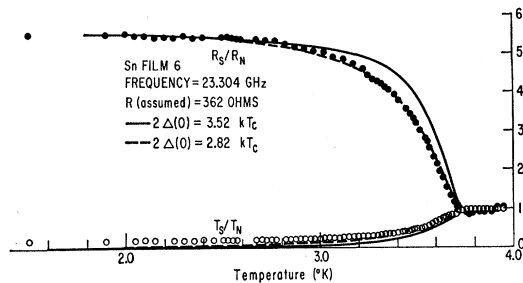
crystal properties. Such films may be composed of small crystallites which have some preferred direction of orientation. Zavaritskii¹⁷ has measured the superconducting energy gap in bulk tin as a function of crystallographic direction using the technique of superconducting tunneling. He found the anisotropy to be quite large with values for $2\Delta(0)$ varying from 2.7 to $4.3 kT_c$.

An attempt was made to fit the data from tin film 6 by varying the value of $2\Delta(0)$ used in the BCS calculations. Figure 9(a) shows the experimental results from tin film 6 compared with theoretical transmission- and reflection-coefficient ratios calculated using $2\Delta(0) = 3.52 kT_c$ (solid line) and $2\Delta(0) = 2.82 kT_c$ (dashed line). Both gaps have the BCS temperature dependence, i.e., $\Delta(T)/\Delta(0)$ is the same in both cases. The agreement between experiment and the curve calculated using $2\Delta(0) = 2.82 kT_c$ is certainly striking though it may be entirely fortuitous. This film was condensed more rapidly than usual and was very thin (note the large value of R_S/R_N at low temperature). The thickness could not be estimated using Eq. (5) because the film developed a clean break during annealing thus making dc measurements impossible.

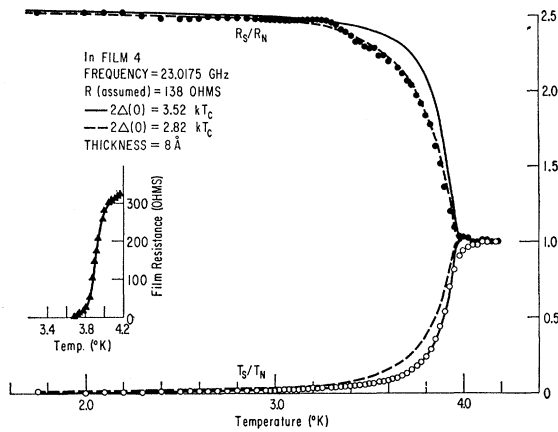
Theoretical curves calculated from conductivities determined from the predicted BCS gap, $2\Delta(0) = 3.52 kT_c$, and from $2\Delta(0) = 2.82 kT_c$ are compared with the experimental data from indium film 4 in Fig. 9(b). The theoretical R_S/R_N from the smaller gap fits the reflection data best, and the theoretical T_S/T_N from the BCS predicted gap fits the transmission data best. No explanation is advanced for this discrepancy.

Structure in T_S/T_N and R_S/R_N

In the previously discussed cases, except for the 71.8 GHz experiment on indium film 2 seen in Fig. 7(d), the microwave frequency was so low that $\hbar\omega < 2\Delta(T)$ at almost all temperatures. Obviously no matter how small $\hbar\omega$, the condition for resonance, $\hbar\omega = 2\Delta(T)$, will



(a)



(b)

FIG. 9. Temperature dependence of the transmission- and reflection-coefficient ratios for tin film 6 and indium film 4. The solid and dashed lines represent T_S/T_N and R_S/R_N calculated from the BCS theory using the parameters given in the legends.

¹⁵ R. W. Vook, J. Appl. Phys. 32, 1557 (1961).

¹⁶ R. H. Blumberg and D. P. Seraphim, J. Appl. Phys. 33, 163 (1962).

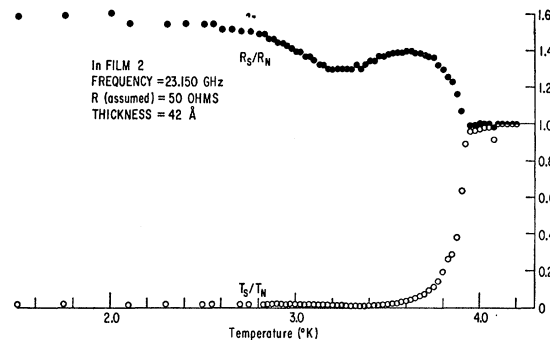


FIG. 10. Temperature dependence of the transmission- and reflection-coefficient ratios for indium film 2 illustrating structure observed in T_S/T_N and R_S/R_N at temperatures well below T_c .

¹⁷ N. V. Zavaritskii, Zh. Eksperim. i Teor. Fiz. 45, 1839 (1963) [English transl.: Soviet Phys.—JETP 18, 1260 (1964)].

be satisfied at some temperature below T_c . For the values of $\hbar\omega$ used in the majority of these experiments this condition for resonance occurs so near T_c that no noticeable effects are observed in T_S/T_N and R_S/R_N because of the rapid increase in $\Delta(T)$ just below T_c . Figures 5, 7(a)-(c), 8, and 9 illustrate this quite clearly.

However, for several films small maxima and minima were observed in the T_S/T_N curves, respectively, at temperatures well below T_c . The results shown in Fig. 10 for indium film 2 are typical. Note that the maximum in T_S/T_N and the minimum in R_S/R_N do not occur at the same temperature. This is consistent with the position of the maximum in T_S/T_N and the minimum in R_S/R_N illustrated in Fig. 6 for case (II) in which $\hbar\omega = 2\Delta(0.927 T_c)$.

Structure in the electromagnetic response of superconducting lead and mercury, consisting of a precursor absorption at energies less than the BCS energy gap, has been observed by Ginsberg and Tinkham,¹⁸ Richards and Tinkham,¹⁹ and Leslie and Ginsberg.²⁰ These authors discuss anisotropy of the BCS gap, states within the gap, and collective excitations as possible explanations of the observed structure.

To account for the type of structure illustrated by Fig. 10, states within the gap would have to be located near the gap edge. Anisotropy of the usual BCS gap would produce a situation very similar to states within the gap. Resonance type behavior, for the low microwave energies used, would occur between BCS gap edges which differ in energy of the order of 10%. However, as pointed out by Ginsberg and Tinkham,¹⁸ the rapid electron scattering in films as thin as the ones investigated in this work, would probably average out any energy gap anisotropy expected from the structure of the Fermi surface.²¹

A satisfactory explanation of the observed structure in the electromagnetic response of superconductors can probably be realized only with more extensive experimental and theoretical investigation.

¹⁸ D. M. Ginsberg and M. Tinkham, Phys. Rev. **118**, 990 (1960).

¹⁹ P. L. Richards and M. Tinkham, Phys. Rev. **119**, 575 (1960).

²⁰ J. D. Leslie and D. M. Ginsberg, Phys. Rev. **133**, A362 (1964).

²¹ L. N. Cooper, Phys. Rev. Letters **3**, 17 (1959).

Transition Temperature

A usual definition for the transition temperature of a superconducting film is "the temperature at which the dc resistance has decreased to one half the residual resistance." For the low microwave frequencies used in these experiments the transition temperature of a superconductor could be defined as the temperature at which T_S/T_N and R_S/R_N differ measurably from 1. The results seen in Fig. 7 show these two definitions of transition temperature to be in good agreement.

V. CONCLUSIONS

The complex conductivities of Mattis and Bardeen² adequately represent the electrical conductivity of thin superconducting films at microwave frequencies. The agreement between experiment and theory is good for a large majority of the cases investigated as illustrated by Figs. 7 and 8. Anomalous behavior occurred for some films even though these films were prepared and measured in the same manner as those that agreed so well with theory. The data for the tunneling films indicates that BCS behavior is being followed by small particles or clumps of metal whose dimensions are approaching the thickness of very thin films. The close agreement between the dc and the microwave transition temperatures illustrated by Fig. 7 indicates a method of measuring the transition temperature of a superconducting film without the use of electrical lead wires and contacts. The influence of the incident microwave power on the transmission and reflection may provide a method of making good critical current measurements. Further experiments in this direction are in progress. A more extensive table of the complex conductivities than that already published²² has been calculated.

ACKNOWLEDGMENTS

The authors are indebted to Professor Rolfe E. Glover, III, for the initiation of this research and his assistance during its early stages. Of considerable use was a table of complex conductivities kindly supplied by J. R. Waldram.

²² P. B. Miller, Phys. Rev. **118**, 928 (1960),