Magnetic Field Dependence of the Surface Impedance of Superconducting Tin at 3 kMc/sec

P. L. RICHARDS*
Royal Society Mond Laboratory, Cambridge, England

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The real and imaginary parts of the surface impedance of superconducting tin have been measured as a function of applied magnetic field by use of a microwave resonator at a frequency of 3 kMc/sec. For most crystalline orientations, the surface resistance and reactance of pure tin were found to decrease with field above 0.85 of the critical temperature but to increase with field at lower temperatures. The negative field dependence, which occurs near the critical temperature, becomes positive when the tin is alloyed with a few tenths of a percent of indium. These results are compared with the Dresselhaus theory, and certain discrepancies are discussed.

I. INTRODUCTION

THE temperature dependence of the microwave surface impedance of superconducting tin at frequencies well below the energy gap is well known from the work of a number of investigators.[1–4] The results of these experiments are best summarized in terms of the two-fluid model of superconductivity which is the appropriate low-frequency limit of the microscopic theory.[5] As the temperature is lowered below the critical temperature T_c, the surface resistance R drops rapidly from the value in the normal state, due to the decrease in the fraction of normal electrons, and vanishes at absolute zero. The surface reactance X also drops rapidly below T_c, but approaches a finite value X_0 at absolute zero, which is determined by the screening of the superconducting electrons alone.

The first measurements of the surface impedance in the presence of an applied magnetic field were made by Pippard[6] using a microwave resonator at a frequency of 9.5 kMc/sec. He found that the surface reactance increased slightly due to the field. This result was in agreement with the thermodynamic theories of Pippard[6] and Bardeen[7] which state that the field would, from the the two-fluid point of view, shift the equilibrium fraction of superconducting and normal electrons in the direction of the normal state and, thus, increase the surface resistance and reactance. His measurements of the surface resistance, however, gave more complicated results. The field caused the surface resistance to decrease at temperatures just below T_c, and to increase at lower temperatures.

Fawcett's[8] measurements of the surface resistance of tin at 36 kMc/sec do not show the negative field dependence found by Pippard at 9.4 kMc/sec. Spiewak's[9] results at 1 kMc/sec, however, show the negative field dependence near the critical temperature in both the surface resistance and reactance. This negative field dependence, which appears from the work of Fawcett, Pippard, and Spiewak to depend strongly on frequency and crystalline orientation, is not well understood.

In order to obtain more information about the frequency dependence and anisotropy of these effects, we decided to make a systematic investigation of the magnetic field dependence of the surface impedance of superconducting tin at 3 kMc/sec as a function of crystalline orientation and temperature.

II. EXPERIMENTAL DETAILS

The microwave techniques employed for our measurements were similar to those used by Pippard[6] and Spiewak.[9] The specimen, in the form of a thin circular cylinder, was hung from a silica halter so as to form the center conductor of a coaxial resonator. The outer conductor was a copper tube small enough to prevent the propagation of any wave guide modes and blanked off a few cm beyond the ends of the specimen to prevent radiation losses. The system resonated at a wavelength slightly longer than twice the length of the specimen.

The microwave power from a klystron oscillator was conveyed through a coaxial cable to a coupling loop about half way along one side of the resonator. The transmitted radiation from a similar loop on the opposite side of the resonator was rectified by a crystal diode and displayed on a galvanometer. The microwave apparatus used was built by Dheer for an investigation of the low-temperature surface impedance of indium and was described more fully elsewhere.[10]

Preparation of Specimens

Because of the anisotropy of the effects to be measured, we decided to use oriented single-crystal specimens for our investigations. The length of the specimens was determined by the chosen resonant frequency to be ap-

* U. S. National Science Foundation Postdoctoral Fellow. Presently at Bell Telephone Laboratories, Murray Hill, New Jersey.
[1] H. London, Proc. Roy. Soc. (London) **A176**, 522 (1940).
[2] A. B. Pippard, Proc. Roy. Soc. (London) **A203**, 98 (1950).
[3] E. Fawcett, Proc. Roy. Soc. (London) **A232**, 519 (1955).
[4] R. G. Chambers, Proc. Cambridge Phil. Soc. **52**, 363 (1956).
[5] J. Bardeen, Phys. Rev. Letters **1**, 399 (1958).
[6] A. B. Pippard, Proc. Roy. Soc. (London) **A203**, 210 (1950).
[7] J. Bardeen, Phys. Rev. **94**, 554 (1954).
[8] E. Fawcett, thesis, University of Cambridge, 1955 (unpublished).
[9] M. Spiewak, Phys. Rev. Letters **1**, 136 (1958); Phys. Rev. **113**, 1479 (1959).
[10] P. N. Dheer, Proc. Roy. Soc. (London) **A260**, 333 (1961).

proximately 4.6 cm. The specimen diameter was limited to less than about 200 μ by sensitivity requirements. The sensitivity of the apparatus is proportional to the skin depth divided by the specimen diameter, so thin specimens were needed for accurate measurements of the changes in surface impedance with applied magnetic field since these were typically of the order of one per cent.

Tin specimens with such small diameters must be supported somehow, and it is convenient to cast them in silica capillaries. Pure tin wires cast in this way do not stick to the silica as they solidify and have bright shiny surfaces. Penetration depth measurements by Pippard[2] on specimens cast in silica capillaries give results very similar to his electropolished specimens. This fact, along with the agreement of our measured values of surface impedance in zero field with the accepted values, leads us to believe that the surface layers of our specimens were representative of the bulk properties of the metal.

Specimens of pure tin oriented along the principal symmetry axes were grown from spectroscopically pure tin obtained from the Vulcan Detinning Company using the following procedure. A short length of silica capillary, sealed at one end, was evacuated and heated to above the melting temperature of tin in a vertical oven. Molten tin was then driven into the capillary by atmospheric pressure air, a seed crystal was connected to the still molten tin, and the capillary was withdrawn vertically from the oven at the rate of about 1 cm/min. Specimens of tin alloyed with varying amounts of indium were also grown by this method.

The single-crystal nature of the specimens was verified by breaking off a small piece from each end, removing the silica in hydrofluoric acid, and determining the crystal orientation using an optical goniometer. All of the specimens grown were single crystals. There appeared to be no tendency to nucleate new crystalline orientations in the capillary. The precise orientations of the specimens was measured to an accuracy of $\pm 2°$ from back-reflections x-ray photographs and is given in Table I in terms of the angles α and β, defined by Fawcett.[3]

All of the specimens, except for No. 9, were vacuum annealed at 190°C; the pure tin specimens for one week; the alloy specimens for two to five weeks depending on their purity.

The ratio $\rho_{295}/\rho_{4.2}$ of the specimen resistivity at room temperature to that at 4.2°K was measured at the completion of the microwave experiments and is shown in Table I. For the pure specimens, these measurements should be taken as a lower bound on specimen quality since the ends of the specimens were damaged considerably when current and potential leads were attached. For the alloy specimens, the resistivity should be less sensitive to cold working, and the measured ratios are in excellent agreement with Doidge's[11] results for the variation of the mean free path in tin alloyed with indium.

[11] P. R. Doidge, Phil. Trans. **A248**, 553 (1956).

TABLE I. Various properties of the experimental specimens.

Specimen No.	Nominal orientation	α	β	Atomic percent indium	$\rho_{295}/\rho_{4.2}$	Diameter in microns
1	[001]	3	44	0	2 900	100
2	[100]	88	0	0	3 500	101
3	[110]	87	45	0	3 500	108
4	[110]	85	44	0	7 500	188
5	[110]	89	42	0.010	1 800	110
6	[110]	86	43	0.030	481	83
7	[110]	89	43	0.20	91	96
8	[110]	87	42	1.00	19	148
9	[110]	87	43	1.00	23	107

Experimental Procedure

At a given value of the reduced temperature $t=T/T_c$, we obtained the ratio R_S/R_N of the surface resistances of superconducting and normal tin by measuring the resonance peak value of the voltage transmission coefficient ζ of the resonator when the specimen was in the superconducting state and when the superconductivity was destroyed by an externally applied magnetic field. Using Pippard's[12] relation $Q/Q_0+K\zeta=1$ between the measured Q, the unloaded Q_0, and ζ, and keeping the coupling constant, one can easily show that

$$\frac{1/\zeta_S(t)-1/\zeta_S(0)}{1/\zeta_N-1/\zeta_S(0)}=\frac{R_S(t)}{R_N}. \tag{1}$$

We approximate $\zeta_S(0)$ by the value measured at the lowest temperature that we could reach (1.2°K) where

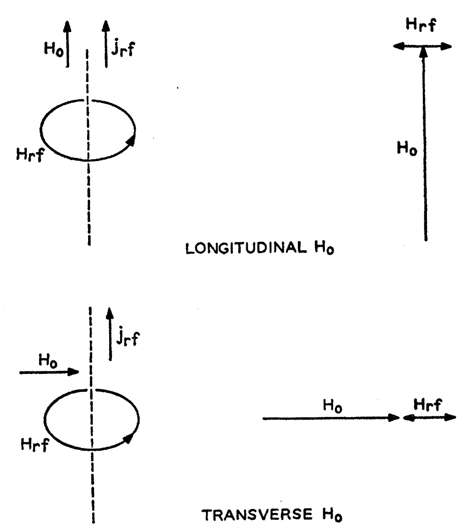

FIG. 1. Field geometry for H_0 longitudinal and transverse to the specimen axis. In both cases the microwave magnetic field H_{rf} circles the specimen and the microwave current j_{rf} flows along the specimen.

[12] A. B. Pippard, Proc. Roy. Soc. **A191**, 370 (1947).

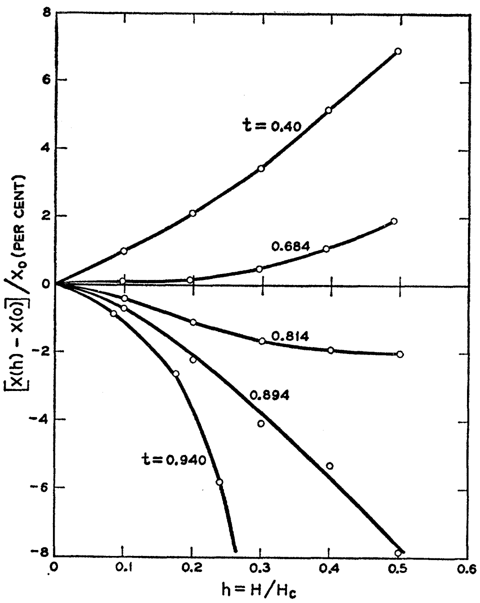

FIG. 2. The surface reactance of a [100] crystal of pure tin (specimen No. 2) in a transverse magnetic field. Due to demagnetizing effects, the specimen enters the intermediate state $h=0.5$.

we assume the losses in the superconducting tin to be negligible. In order to determine the field dependence, the changes in the resonance peak values of ζ were noted as transverse and longitudinal magnetic fields were switched on and off. Using (1) we then calculated values of $[R_S(h)-R_S(0)]/R_N$, where $h=H/H_c$ and H_c is the critical field at temperature t.

In this analysis we have assumed that R_N is independent of temperature and magnetic field over the range of our measurements. Experimentally, changes in R_N of the order of one per cent were found for our pure tin specimens, but they will be ignored, as their effect on the calculated quantities is small compared with the errors in the field dependence of ζ_S.

Changes in the surface reactance X with field or temperature appear as changes in the resonant frequency of the cavity. In order to measure the small changes with field, the oscillator frequency was adjusted so that the output of the resonator was at three-quarters of the peak output in the presence of the magnetic field (at this level the change of output with frequency is most rapid). Observations were then made of the changes in output as the field was turned off. The measurement was then repeated on the other side of the resonance. These changes in output, together with the known Lorentzian line shape and measured width of the resonance in zero field, are sufficient to calculate the shift of the resonance frequency with the applied magnetic field. As in the case of the surface resistance, the frequency shifts for each specimen were measured as a function of magnetic field and temperature.

For display purposes, it was decided not to convert the measured frequency shift directly into changes in X, but to normalize them with the shift that corresponds to X_0, the value of the superconducting surface reactance at absolute zero. In order to obtain X_0, the difference in resonance frequency between the superconducting and normal states was measured as a function of temperature for each specimen. This difference was then plotted as a function of $(1\text{-}t^4)^{-\frac{1}{2}}$. The experimental points fell very nearly on a straight line whose slope was taken as the frequency shift corresponding to X_0.

The basis for this procedure is the empirical law,[13] $\lambda=\lambda_0(1-t^4)^{-\frac{1}{2}}$, for the temperature dependence of the penetration depth λ. At sufficiently low temperatures that the surface resistance of the superconductor is negligible, X is proportional to λ so the slope of our line should give a satisfactory value of the frequency shift corresponding to X_0. Unfortunately, the data were taken over the range $1.1<(1-t^4)^{-\frac{1}{2}}<1.8$ over which R_S cannot be neglected in the pure tin specimens. For these, a curvature in the data was noticed, especially in the high temperature range. As it is difficult to estimate an accurate correction for this curvature, it was decided to use the slope of a best-fit straight line over the range $1.1<(1-t^4)^{-\frac{1}{2}}<1.5$ as the normalizing factor. The value of X_0 so determined is an underestimate, but its use should not affect the important qualitative features of the field dependence data.

The geometrical factor[2] which converts changes in resonance frequency to changes in surface reactance or penetration depth was calculated, neglecting end effects, from the diameter of the resonator and the diameters of the specimens given in Table I. We found the value 4.7×10^{-6} cm for the penetration depth at absolute zero in pure [110] specimens of tin. The penetration depth increased linearly with indium concentration to the value 7.5×10^{-6} cm for specimen No. 8 with one per cent indium. Specimen No. 9, (also with one per cent indium, but not annealed) had a penetration depth of 1.5×10^{-5} cm which, if we extrapolate using our linear dependence of λ on purity, would correspond to an annealed specimen with roughly 3.7% indium.

Our results for the penetration depth in pure tin are about 10% below those of Pippard[14] but very nearly coincide with his results for a specimen containing one per cent indium. From the considerations given above, it is not surprising that our values of the penetration depth in pure specimens should be too low.

Two more features of the experimental procedure should be mentioned before we proceed to an examination of the results. We measured the critical temperatures of our specimens (by measuring R_S/R_N in the vicinity of T_c) as a function of the microwave power dissipated in the specimen. Extrapolating to zero microwave power, we found $T_c=3.72$°K on the 1958 tempera-

[13] D. Shoenberg, *Superconductivity* (Cambridge University Press, Cambridge, England, 1952), 2nd ed.
[14] A. B. Pippard, Proc. Roy. Soc. (London) **A216**, 547 (1953).

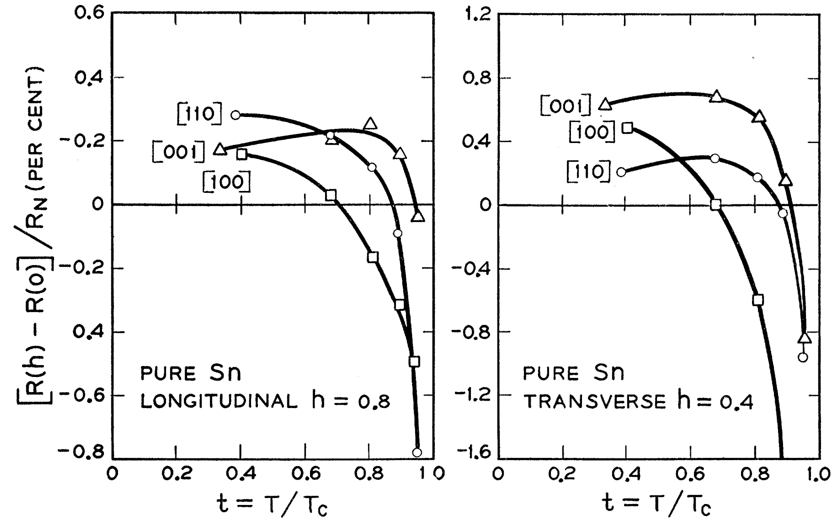

FIG. 3. The surface resistance of pure tin specimens in longitudinal and transverse magnetic fields.

ture scale[15] for our pure specimens and a decrease in T_c with indium concentration very similar to that found by Doidge.[11] After correcting for heating, we discovered that the measured fractional changes in surface impedance with magnetic field were essentially independent of the microwave power level.

The second check made on each specimen was to observe the resonance amplitude in the superconducting state as a critical magnetic field was switched on and off for the first time. None of the specimens used showed any evidence of flux trapping. The Meissner effect appeared to be perfect in each case.

III. EXPERIMENTAL RESULTS

We measured the fractional change in surface resistance $[R(h)-R(0)]/R_N$ and surface reactance $[X(h)-X(0)]/X_0$ with magnetic field for longitudinal and transverse field orientations as shown in Fig. 1, at five different values of the temperature for each of our nine specimens. As an example of the data we show, in Fig. 2, the transverse field dependence of the reactance of specimen No. 2. The qualitative features of these curves are representative of most of our data. The magnetic field decreases X at temperatures near T_c but increases it at lower temperatures. The field dependence is nearly quadratic for small h, but becomes more complicated for larger fields. Because of demagnetizing effects, the specimen enters the intermediate state when $h=0.5$ and, for larger values of h, $X(h)$ rises rapidly toward the normal state value. In a longitudinal field similar behavior occurs at $h=1$. It should be kept in mind that because of the temperature dependence of H_c, the value of the field that corresponds to a given h is temperature dependent.

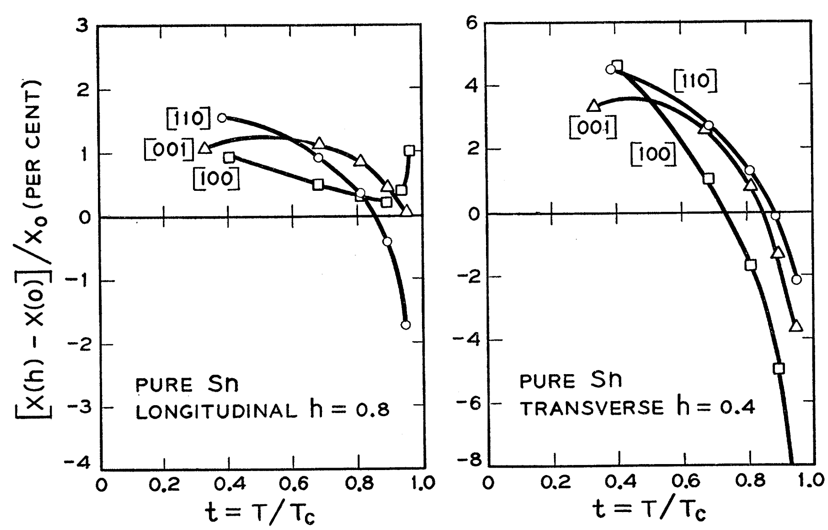

FIG. 4. The surface reactance of pure tin specimens in longitudinal and transverse magnetic fields.

[15] H. van Dijk and M. Durieux, Physica 24, 920 (1958).

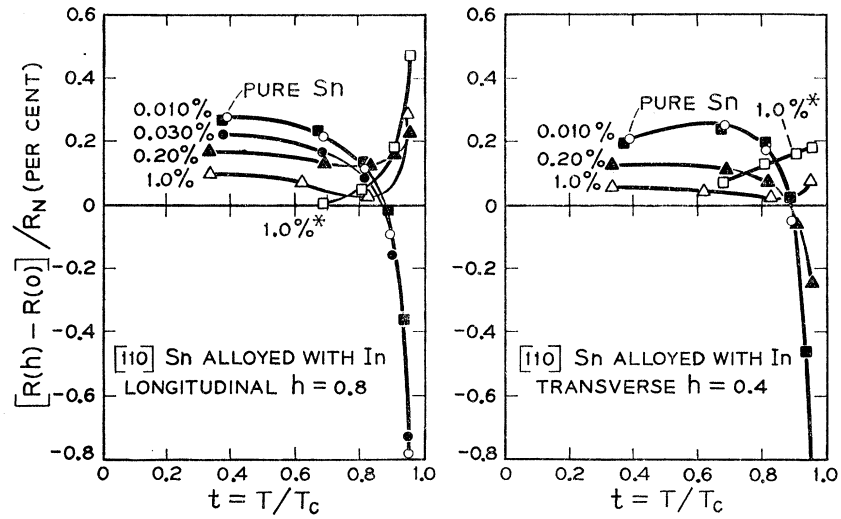

FIG. 5. The surface resistance in longitudinal and transverse magnetic fields of tin specimens alloyed with small amounts of indium. All of the specimens were thoroughly annealed except for the one marked 1.0%* which corresponds roughly to an annealed specimen with between 3 and 4% indium.

Rather than to attempt to compare the qualitative features of 36 figures similar to Fig. 2, we have replotted the data as a function of temperature at 0.8 of the field at which normal state or intermediate state effects begin to appear, that is, at $h=0.8$ for the longitudinal field data and at $h=0.4$ for the transverse field data. These combined plots are given in Figs. 3 and 4 for the pure tin specimens and in Figs. 5 and 6 for the alloy specimens.

Our measurements on pure tin show a variety and anisotropy similar to that observed by Spiewak[9] at 1 kMc/sec. The results for the resistance and reactance of a [100] crystal (specimen No. 2) in a transverse field are qualitatively similar to the corresponding curves for for Spiewak's specimen Sn-1, which was oriented about 10° from the [100] axis. The longitudinal field results, however, differ markedly. This indicates either a complicated frequency dependence or an exceptionally strong anisotropy in the longitudinal field case. Spiewak's other specimens were oriented too far from the symmetry axes to allow a direct comparison.

The anisotropy of the field dependences in pure tin is very large for both field directions. The [110] and [100] specimens show qualitatively different results indicating that the anisotropy is nontensorial. Since there is no convincing evidence for nontensorial anisotropy of X_0 in tin, our results indicate that the normal electrons must play an important role in the observed effects.

Because of the Meissner effect, the local magnetic field strength at the surface of the specimen varies between zero and twice the applied field in the transverse geometry. Thus the measured field dependence of the surface impedance is an average of the local dependence corresponding to values of H_0 in this range. One can easily show from the known field distribution around the specimen that, if the field dependences are quadratic and if the specimen is isotropic with respect to rotations about its axis, then the measured average

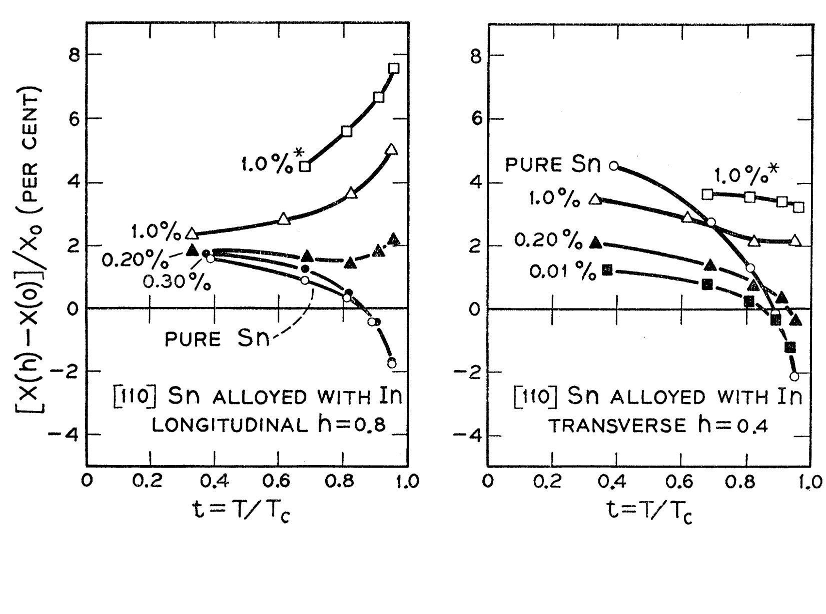

FIG. 6. The surface reactance in longitudinal and transverse magnetic fields of tin specimens alloyed with small amounts of indium. All of the specimens were thoroughly annealed except for the one marked 1.0%* which corresponds roughly to an annealed specimen with between 3 and 4% indium.

field dependence is still quadratic but with half the value at $h=0.5$ that the local field dependence has at a local field of $h=1$.

Measurements of the azimuthal anisotropy made by rotating the specimen in a transverse magnetic field revealed no anisotropy for our [001] and [100] specimens, but an anisotropy similar to that shown in Fig. 7 for all of our [110] specimens. Thus, because of the small size of the azimuthal anisotropy found and the approximately quadratic character of the field dependence, we can make an approximate correction for demagnetizing effects by doubling the field dependences and the magnetic fields in the transverse case. When this is done, we find that the field dependences of both R and X for pure tin are about four times as large in the transverse geometry as in the longitudinal.

This result should be compared with an argument due to Ginsburg and Landau[16,17] which predicts that if the superconductor is able to follow the oscillations of the microwave magnetic field, then the apparent change in penetration depth, λ, (and thus at least the low temperature changes in X) should be three times as large in our transverse field where H_{rf} is parallel to H_0 as it is in our longitudinal case where H_{rf} is perpendicular to H_0 (see Fig. 1). This apparent agreement with our experimental results should be treated with caution since when impurities are added one would expect the superconductor to relax more rapidly to the instantaneous value of the microwave field. Yet we find the transverse field effects only about twice as large as the longitudinal effects for our less pure alloy specimens.

IV. REVIEW OF CURRENT THEORY

A theory of the magnetic field dependence of the superconducting surface impedance proposed by Dresselhaus and Dresselhaus[18] is based on the bending of the orbits of the normal electrons in the superconducting state by a static magnetic field. The Dresselhaus theory is sufficiently flexible that, by using the effective mass of the normal electrons in the superconducting state as an adjustable parameter, a moderately good fit to some of Spiewak's data can be obtained.[18] A similar partial fit can be made to our results for pure tin. In order to do this, however, one must assume that the effective mass (or more exactly the effective mass times the Fermi velocity) has the free electron value when $H_0=0$, but decreases by a factor of nearly 10^3 when a magnetic field of a few tens of oersted is applied. The orbit diameter of electrons with such small values of m^*v_f would approach the diameter of our smaller specimens in a field of a few oersteds. Thus one might hope

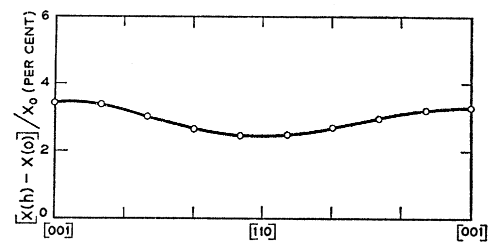

FIG. 7. Variation of the field dependence of the surface reactance of a typical [110] specimen with the azimuthal angle of the transverse magnetic field.

to see a size effect in the field dependence of the surface impedance near T_c. A careful comparison of the results from our pure tin specimens Nos. 3 and 4 which have the same orientation, but differ by nearly a factor to two in diameter, does not reveal any observable size effect. Only the more accurate data for the thinner specimen, No. 3, are given in Figs. 3–6.

Our results for tin alloyed with indium are in clear disagreement with the predictions of the Dresselhaus theory. Figures 5 and 6 show that the negative field dependences of R and X disappear when indium is added. By using our experimental values of the penetration depth $\lambda(t)$ and normal state mean free path l, and by assuming that the coherence length in pure tin has the value[19] $\xi_0=2.1\times10^{-5}$ cm and that $1/\xi=1/\xi_0+1/l$, we have estimated the values of the parameters ξ/λ and l/λ at the temperature $t=0.95$ and at the indium concentration for which the negative effects disappear. The results given in Table II show that ξ/λ does not change

TABLE II. Experimentally determined [110] specimen parameters for the indium concentrations at which the field dependence of R and X becomes positive at $t=0.95$. Values are given for both field orientations and (for the sake of comparison) for pure tin.

	ξ/λ	l/λ
R in longitudinal H_0	1.6	21
X in longitudinal H_0	1.6	23
R in transverse H_0	0.9	2.2
X in transverse H_0	1.3	5.6
Pure tin at $t=0.95$	1.8	350

much from the value in pure tin at $t=0.95$, so that l/λ is perhaps a more significant parameter. We can say that when enough indium is added so that l/λ is of the order of 10, that is, when the extreme anomalous limit of the skin effect theory for the normal electrons begins to fail, the negative field dependences of R and X disappear. The Dresselhaus theory, on the other hand, predicts that as the limit of the classical skin effect is approached, the field dependence of R becomes positive, but that of X remains negative.

We feel that it would not be profitable to attempt a detailed fit of our data to the Dresselhaus theory in its

[16] V. L. Ginsburg and L. D. Landau, J. Exptl. Theoret. Phys. (U.S.S.R.) **20**, 1064 (1950).

[17] A. B. Pippard, *Advances in Electronics and Electron Physics*, edited by L. Marton (Academic Press, Inc., New York, 1954), Vol. 6, pp. 1–45.

[18] G. Dresselhaus and M. Spiewak Dresselhaus, Phys. Rev. **118**, 77 (1960); Phys. Rev. Letters **4**, 401 (1960).

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

present state. We will therefore, conclude this paper with a discussion of several other theoretical ideas proposed as an interpretation of these effects.

Bardeen[5] has suggested that the negative field dependence of the surface impedance may be associated with a counterflow of normal and superconducting fluids perpendicular to the surface in the penetration region. Such a disturbance of the equilibrium, which is explicitly neglected in the Dresselhaus theory, would presumably be sensitive to the total magnetic field vector and so should be more pronounced in the transverse field geometry where the magnitude of the total magnetic field vector oscillates at the microwave frequency than in the longitudinal case where its magnitude is nearly constant (see Fig. 1). From Figs. 3 and 4 we see that the negative field dependence of X is indeed more pronounced in the transverse geometry, although the same cannot be said for R. If we consider all of our pure tin specimens (including two with unknown crystalline orientation which were partially studied during the preliminary testing of the apparatus) and all three of Spiewak's specimens, we find that $[X(h)-X(0)]/X_0$ becomes negative for two of six specimens in the longitudinal orientation, but for every one of the eight specimens measured in the transverse orientation. This is a surprising result from the point of view of the Dresselhaus theory which makes no qualitative distinction between the two field orientations. It suggests that a disturbance of the equilibrium fraction of superconducting and normal electrons, such as that suggested by Bardeen, might play an important role in the negative field dependence of X. The few instances of a negative change in X for a longitudinal field could be explained by a coupling to the two fluids by the anisotropy of the Fermi surface.

The thermodynamic models of Pippard[6] and Bardeen[7] predict that the equilibrium result of a static magnetic field is to shift the superconductor in the direction of the normal state and thus to increase both R and X. It is therefore difficult to see how a simple relaxation of the equilibrium toward the instantaneous, total magnetic field could produce the negative changes in R and X measured at microwave frequencies. We are thus led to consider the possibility of resonance phenomena in the penetration layer. In order to investigate this idea, Pippard[20] has made a semiquantitative survey of the frequency dependence of the magnetic field dependence of the surface impedance using the data of Fawcett,[3] Pippard,[6] Spiewak,[9] and the present work. He finds that the phase angle of a vector representing the change with magnetic field of the complex superconducting skin depth varies with frequency much as the phase of the complex response of an electrical circuit varies when passing through a resonance. In the superconducting case, however, if one uses the thermodynamic result to determine the phase at zero frequency, the phase change is through nearly 360°, in contrast to the 180° found for a conventional resonance, or the 90° phase change associated with a simple relaxation. Pippard's arguments are necessarily approximate since they rest on comparisons of data ignoring the large crystalline anisotropy, but they are very suggestive and stress the importance of obtaining data at lower frequencies to test the validity of the thermodynamic arguments.

V. CONCLUSIONS

Our measurements of the magnetic field dependence of the surface impedance of superconducting tin at 3 kMc/sec give results that are fully as complicated as those found by Spiewak at 1 kMc/sec. We find discrepancies between our results and the Dresselhaus theory, especially for the behavior of alloy specimens and the dependence on magnetic field orientation, which indicate that the theory of these effects is not entirely satisfactory. It seems possible that the observed effects result more from a disturbance of the equilibrium fraction of superconducting and normal electrons rather than from any changes in the properties of the normal electrons. Further experimental work at much lower frequencies would be helpful in clearing up this point.

ACKNOWLEDGMENTS

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[20] A. B. Pippard, *Proceedings of the Seventh International Conference on Low-Temperature Physics*, 1960 (University of Toronto Press, Toronto, 1961), pp. 320–327.