

*DEPENDENCE OF THE DEPTH OF PENETRATION OF THE MAGNETIC FIELD IN A SUPERCONDUCTOR ON THE MAGNETIC FIELD STRENGTH*

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The dependence of the depth of penetration  $\delta$  of the magnetic field in a superconductor on the field was measured in 10 samples placed in the coil of the resonant circuit of an oscillator operating at 2 Mc/sec. The change in frequency accompanying the application of the field was measured accurate to 0.005 cps, corresponding to a change in  $\delta$  of about  $10^{-9}$  cm.

THE depth of penetration of a magnetic field in an isotropic superconductor is defined as the ratio of the magnetic flux in a surface layer of the superconductor to the value of the magnetic field on this surface, i.e.,

$$\delta = \int_0^{\infty} H(z) dz / H(0)$$

(it is assumed that the permeability  $\mu = 1$ ). This quantity can be considered independent of the magnetic field only in first approximation, at small values of  $H/H_c$ , when the equations of superconductor electrodynamics are assumed linear. In general, we can put

$$\delta(H) = \delta(0)[1 + \alpha(H/H_c)^2 + \beta(H/H_c)^4 + \dots], \quad (1)$$

since  $\delta$  is obviously an even function of  $H$ .

The question of the magnitude and the character of the nonlinear effects in real superconductors has not yet been sufficiently clarified both in its experimental and theoretical aspects. At temperatures sufficiently close to  $T_c$ , the phenomenological theory of Ginzburg and Landau<sup>1</sup> leads to the relation

$$\alpha = \frac{\kappa(\kappa + 2\sqrt{2})}{8(\kappa + \sqrt{2})^2}, \quad \kappa = \frac{\sqrt{2}|e_{\text{eff}}|}{\hbar c} H_c \delta^2(0). \quad (2)$$

Using the method indicated in reference 1, expressions can also be derived for the succeeding expansion coefficients in (1). The exact expression for  $\beta$  is quite cumbersome. Approximately, when  $\kappa \ll 1$ ,

$$\beta = 4.7 \cdot 10^{-2} \kappa^2 (1 - 5.1\kappa + 7\kappa^2) \text{ and } \beta \ll \alpha. \quad (3)$$

Gor'kov<sup>2</sup> has shown, starting with the modern microscopic theory of superconductivity,<sup>3,4</sup> that  $e_{\text{eff}}$  in (2) should be taken equal to twice the electron charge, and the validity of relations (2) and

(3) should be restricted both by the condition  $(T_c - T)/T_c \ll 1$  and by the condition  $\delta \gg \xi_0$ , where  $\xi_0$  is the nonlocality parameter.<sup>3</sup> For tin this condition corresponds approximately to  $T_c - T < 0.1^\circ$  (see reference 5).

There exists apparently no theoretical analysis of the nonlinear effects in the region where the local relations of the Ginzburg and Landau theory are not applicable.

The first attempts at an experimental investigation of the dependences for tin and mercury were made quite long ago, simultaneously with the first measurements of  $\delta$  in weak fields. For mercury, Laurman and Shoenberg<sup>6</sup> found the changes in  $\delta$  to be beyond the limits of the sensitivity of their method, corresponding to  $\alpha < 0.05$ .

The function  $\delta(H)$  for tin was investigated<sup>7,8</sup> by low-frequency ( $\nu = 4$  cps) modulation of the sample temperature. It was noted that polycrystalline mechanically worked and annealed samples exhibit strong and complicated dependences of  $\delta$  on the applied field. In single-crystal tin samples<sup>8</sup> the increments of  $\delta$  were several times smaller, and the  $\delta(H)$  plot became approximately parabolic. Still the value  $\alpha = 0.06$  for  $2.4^\circ \leq T \leq 3.67^\circ$ , determined from these experiments, exceeded appreciably the theoretical value  $\alpha = 0.025$ , obtained by substituting in (2) the expression

$$\delta(0) = 2.5 \cdot 10^{-6} (1 - T/T_c)^{-1/2} \text{ cm}, \quad (4)$$

which is valid near  $T_c$ .<sup>6,8-10</sup>

To ascertain the reasons for this discrepancy, we have decided to carry on further investigations on a large number of samples, using a more accurate and more sensitive radio-frequency procedure for the measurements.

Pippard<sup>9,11</sup> has demonstrated that the use of high frequencies for the investigation of the depth of penetration has considerable methodological advantages over low-frequency statistical measurement methods. In this case the errors due to the presence of an included normal phase in the sample are considerably reduced (if the depth of penetration of the field in the normal phase is less than the dimensions of these inclusions). The sensitivity of the apparatus to changes in the depth of penetration can also be appreciably increased by raising the frequency of the measuring field, since the reactive component  $X$  of the surface impedance of the sample is connected with the depth of penetration, at not too high frequencies, by the formula

$$X = 4\pi c^{-2} \delta \omega \quad (5)$$

and increases consequently as the frequency. The closer the temperature of the sample is to  $T_C$ , the more stringent is the limitation that should be imposed on the frequency in order to satisfy the equality (5).

Theoretical estimates for the frequency interval in which relation (5) can be used for the determination of the statistical value of  $\delta$  have been made by Abrikosov, Gor'kov, and Khalatnikov.<sup>12</sup> However, our own choice of the operating frequency is based only on the experimental data, which call for more stringent limitations.

From this point of view, the 9400 Mc/sec frequency used by Pippard<sup>13</sup> in the investigation of the dependence of the impedance of tin on the field is too high. In this case the linear dependence of  $X$  on  $\delta$  is noticeably violated even at temperatures above 3°K.<sup>11</sup> When  $T_C - T = 0.1^\circ$ , the difference between the static value<sup>6</sup> of  $\delta(0)$  and the value  $\delta_\omega(0)$  as given by formula (5) amounts to approximately 40% of  $\delta(0)$ . A reliable determination of the static value of  $\alpha$  near  $T_C$  is impossible in this case.

In Pippard's measurements<sup>9</sup> at 1200 Mc/sec with no constant field, a value of approximately 10% was obtained for  $[\delta(0) - \delta_\omega(0)]/\delta(0)$  at  $T_C - T = 0.1^\circ$ . A still lower frequency should make the difference  $\delta(0) - \delta_\omega(0)$  increase even more rapidly, since it follows from general considerations that at low frequencies  $\omega$  we have  $\delta - \delta_\omega \sim \omega^2$  (see reference 14).

We note that an analogous frequency dependence, proportional to  $\omega^2$  was actually found by Sturge<sup>15</sup> for the active component of the surface impedance of tin at frequencies from  $10^3$  to  $10^2$  Mc/sec.

In the development of the measurement procedure, we decided on a frequency near 2 Mc/sec,

which is certainly sufficiently low in the investigated temperature region, where  $T_C - T > 0.01^\circ$ . On the other hand, it was possible to attain at this frequency an accuracy and sensitivity which were fully adequate for our purposes.

## THE METHOD

**Construction of the instrument.** The heart of the measuring apparatus were two identical oscillator circuits using 06-P2B low-power pentodes placed in a Dewar vessel with liquid helium (the parts kept at low temperature are separated in Fig. 1 by a dashed line). The two oscillators together dissipated in the helium approximately 0.05 w. The investigated sample, in the form of a cylinder with pointed ends (diameter 8–9 mm,

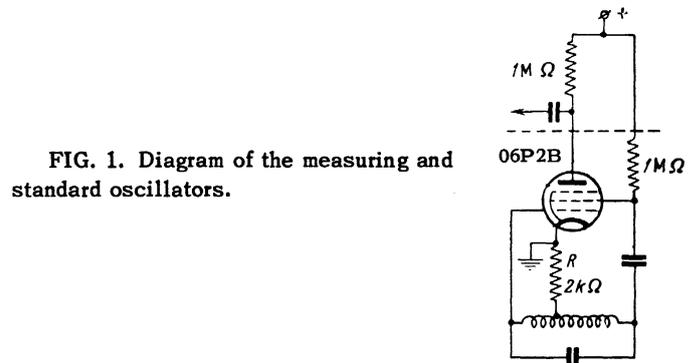


FIG. 1. Diagram of the measuring and standard oscillators.

length of cylindrical part 40 mm, overall length 60 mm), was placed in the coil (approximately 20 mm long) of the tank circuit of one of the oscillators (Fig. 2). The coil was placed in the homogeneous magnetic field of solenoid A (Fig. 3). The earth's field was compensated for during the experiment with an accuracy of  $\sim 0.01$  oe. The change in the surface impedance of the sample due to the application of the external magnetic field changed the frequency  $f_1$  of the first oscillator. The second oscillator served as a frequency standard. Both oscillators were connected to a common power pack through dependable decoupling filters. In these Hartley oscillators the B voltage was applied to the screen grids of the pentodes, and the signal was picked off the plates, to reduce the coupling between the output and the tank circuits. Small resistances ( $R$  in Fig. 1) were inserted in the cathode circuits to reduce the frequency shift due to variation of the filament current. The second oscillator and the tube of the first oscillator were shielded against external magnetic fields by lead superconducting shields. The frequency  $f_0$  of the second generator could be varied over a small range during the time of the experiment by

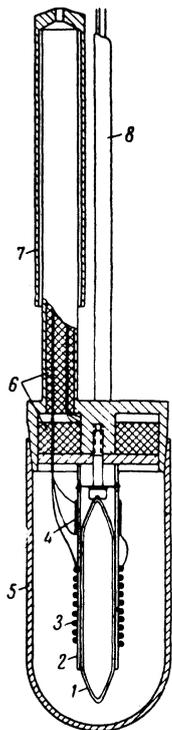


FIG. 2. Section through the measuring oscillator. 1 - sample in ampule, 2 - quartz form of the tank circuit, 3 - coil winding - 100 turns of copper wire (0.15 mm dia.), 4 - mica capacitors of the tank circuit, 5 - copper vacuum-tight screen with lapped seal, 6 - foamed styrol heat insulation between the tube and the sample, 7 - lead shield, 8 - tube for vacuum and wire leads.

means of a lead plunger traveling in the coil of its own tank circuit.

The signals from both oscillators were fed to a mixer M (Fig. 3), which separated the difference frequency  $f_1 - f_0$ . The value of the difference frequency was set equal to approximately 100 cps. The value of  $f_1 - f_0$  was adjusted and roughly measured by means of an ICh-6 frequency meter feeding an ÉPP-09 automatic recorder, and by means of an oscillograph with a sweep signal from a ZG-12 audio oscillator. For exact measurement of the frequency, the sinusoidal signal was converted in the shaping network S into a sequence of pulses, the interval between which was increased by a factor  $k_1$  in a PS-64 scaler (in most experiments,  $k_1 = 64$ ). The interval between pulses was measured with a chronograph comprising a crystal oscillator G, an electronic switch SW which passed a signal of frequency  $\varphi = 10^5$  cps from the crystal oscillator in the interval between two pulses, and PS-10000 counters, which counted

the number of cycles in the interval between the pulses. The measurements were made with the magnetic field of solenoid A alternately switched on and off several times (usually 16 times); the number of cycles counted with the field off,  $N_1$ , was summed by one PS-10000 counter, while the number of cycles with the field on,  $N_2$ , was counted by the second counter. Relay Rel was used to actuate the devices and to apply the magnetic field, and the number of switchings  $k_2$  was set by means of a supplementary PS-64 scaler.

**Calibration of the instrument.** The frequency shift  $\Delta f_1$  due to application of the magnetic field could be determined from the relation  $\Delta f_1 = k_1 k_2 \varphi (N_1 - N_2) / N_1 N_2$ . The accuracy of the measurement of  $\Delta f_1$  was determined by the stability of the oscillators and by the duration of the measurements, and amounted to approximately 0.005 cps. To calibrate the instrument, the frequency  $f_1$  was determined for lead samples of different radii r. The same measurements were repeated with aluminum samples in normal state, and the results were in agreement. These data were used to calculate, accurate to 4 or 5%, the value of  $dr/df_1$  as a function of the radius of the sample. For the investigated tin samples,  $dr/df_1 = (3 - 4) \times 10^{-7}$  cm/cps. Thus, depth-of-penetration changes on the order of  $10^{-9}$  cm could be detected.

**Control experiments.** The influence of the field on the oscillation frequency in the absence of a sample were checked between individual measurements. To reduce this influence, we had to make the tank-circuit coil of specially prepared copper wire, the surface of which was free of iron. All the joints in and near the tank circuit were made with a solder which was not superconducting in the operating frequency range (60% Bi, 40% Cd). With these precautions, the absolute frequency shift due to application of a field ranging from 0 to 100 oe could not exceed 0.02 - 0.03 cps. This effect could appreciably reduce the measurement accuracy only at temperatures below 3° K. It can be assumed that the frequency shift was due essentially to mechanical displacement of the parts of the instru-

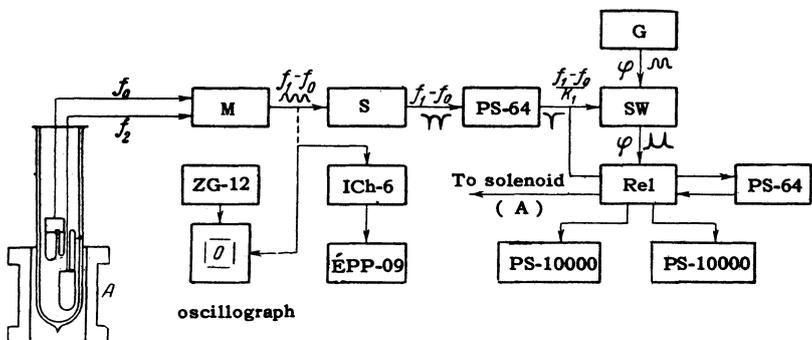


FIG. 3. Block diagram of the measuring apparatus.

ment due to switching on the magnetic field, since the magnitude of the observed displacement was not consistently reproducible.

**Data reduction.** In determining  $\delta(H)$  from our data it is necessary to take it into account that if  $\delta$  depends on the magnetic field Eq. (5) is not applicable directly, for it is necessary then to allow for the periodic variation of  $\delta$  induced by the measuring field. Assuming the oscillation frequency to be sufficiently low, we can write down, accurate to the fifth power of the field, the instantaneous value of the magnetic flux through the surface layer of the sample per unit length of its circumference as

$$\Phi = H\delta(0)(1 + \alpha H^2/H_c^2 + \beta H^4/H_c^4), \quad (6)$$

where  $H$  is the sum of the constant and measuring fields, i.e.,  $H = H_0 + H_1 \sin \omega t$ . Substituting this expression in (6), we should single out the terms that oscillate at a frequency  $\omega$  and depend on  $H_0$ , for only these terms can change the oscillation frequency. In most of our experiments,  $H_0 \parallel H_1$ . Then, putting  $H_0/H_c = h$  and  $H_1/H_c = h_1$ , we obtain

$$\Phi_\omega = \delta(0) \left[ \left( 3\alpha + \frac{15}{2} \beta h_1^2 \right) h^2 + 5\beta h_1^4 \right] H_1 \sin \omega t. \quad (7)$$

The measuring field  $H_1$  was measured in separate experiments under different conditions, and was set at 0.1–0.2 oe during the measurements. The quantity  $15\beta h_1^2/2$  amounted to a fraction of one percent of the total coefficient of  $h^2$ , and could therefore be neglected. The effective increase in  $\delta$ , determined experimentally as  $\Delta_{\text{eff}}\delta = (d\delta/df_1) \times \Delta f_1$ , was expressed thus in terms of  $\alpha$  and  $\beta$  as

$$\Delta_{\text{eff}}\delta = \delta(0)(3\alpha h^2 + 5\beta h^4). \quad (8)$$

The value of  $\alpha$  could therefore be determined from the initial slope of the  $\Delta_{\text{eff}}\delta$  vs.  $h^2$  curve.  $\delta(0)$  was calculated here from (4).

**Samples.** The characteristics of the investigated samples are listed in the table. At the preliminary stage of the investigation we used samples grown in glass ampules, which were removed from the samples prior to the investigation. One such sample is numbered 1 in the table. The remaining samples were placed in the coil in quartz ampules, the internal surface of which was coated with a thin layer of lampblack to prevent the tin from sticking during casting. The ampules removed from the samples were placed in the coil during the control experiments. The crystal orientation of the samples was determined from the etch figures. Samples 3 and 4 consisted of several crystals, turned 1–2° relative to each other. These

Sample number	$\frac{\rho(4,2^\circ\text{K})}{\rho(20^\circ\text{C})} \cdot 10^5$	Impurities,* 10 <sup>-4</sup> %	$\varphi$ **, deg	$\psi$ ***, deg
1	4	8	65	30
2	1	1	90	0
3	1	1	40	30
4	1	1	75	0

\*Impurity content estimated according to reference 16.

\*\* $\varphi$  – angle between the axis of the sample and the [001] axis.

\*\*\* $\psi$  – angle between the projection of the sample axis on the (001) plane and the [100] axis.

crystals apparently came from a single seeding under conditions where the tin was considerably supercooled.

## RESULTS

The general form of the function  $\Delta f_1(H)$  is shown for one of these samples in Fig. 4. The curve was obtained with the ICh-6 frequency meter and an automatic recorder. The instant when superconductivity is destroyed is noted from the sharp kink on the curve. Superconductivity was sometimes destroyed at fields somewhat higher than  $H_c$ , and consequently the values of the critical field were determined by observation of the reverse transition from the intermediate to the superconducting state.

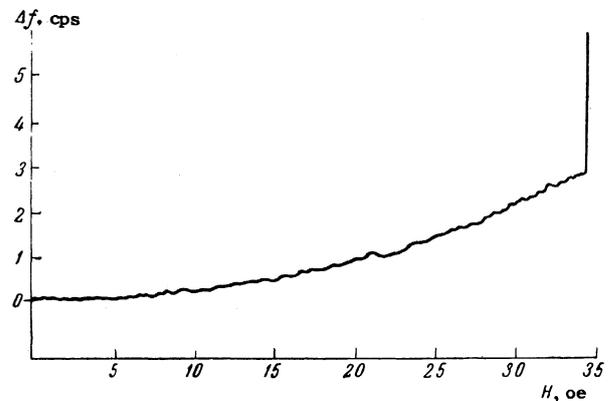


FIG. 4

Figure 5 shows the results of the measurements of  $\Delta_{\text{eff}}\delta$  for sample 3, made with the aid of a chronograph. The dependence of  $\Delta_{\text{eff}}\delta$  on  $h$  was close to parabolic, although noticeable deviations from a parabola were observed at large values of  $H$ , particularly at low temperatures. The initial slopes of the curves were determined by using two points carefully measured at small values of  $h^2$ . The dispersion in repeated measurements usually did not exceed 2 or 3%. The values of

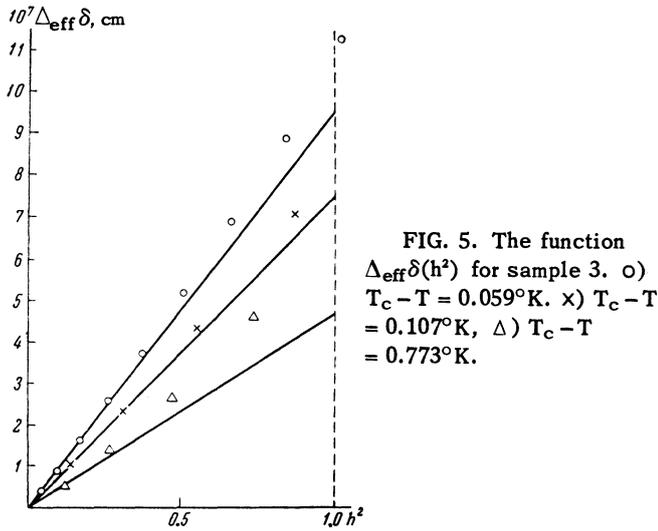


FIG. 5. The function  $\Delta_{\text{eff}}\delta(h^2)$  for sample 3.  $\circ$ )  $T_c - T = 0.059^\circ\text{K}$ .  $\times$ )  $T_c - T = 0.107^\circ\text{K}$ ,  $\Delta$ )  $T_c - T = 0.773^\circ\text{K}$ .

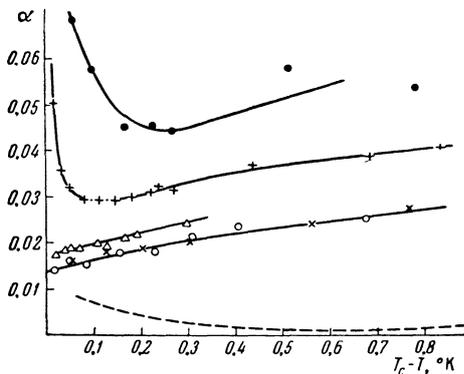


FIG. 6. The function  $\alpha(T)$ .  $+$ ) sample 1;  $\Delta$ ) sample 2;  $\times$ ) sample 3;  $\circ$ ) sample 4;  $\bullet$ ) sample 4 after removal of the ampule.

$\alpha(T)$ , calculated for all samples listed in the table, are shown in Fig. 6. Measurements on sample 1 led to values of  $\alpha$  which increased rapidly as  $T \rightarrow T_c$ . Further experiments showed, however, that this temperature variation of  $\alpha$  was due to secondary causes. Although the tin surface was carefully protected against scratches, the sample might have been slightly bent during the installation and cooling. Samples 2–4, protected by ampules against possible damage, yielded considerably smaller, nearly equal values of  $\alpha$ , which tended to a finite limit as  $T \rightarrow T_c$ . After the ampule was taken off sample 4, the values of  $\alpha$  for this sample increased sharply and the  $\alpha(T)$  curve assumed the same form as for sample 1. It must be noted that the anomalous behavior of the deformed specimens was not due to the incompleteness of the Meissner effect, for the results with all specimens were practically independent of the magnitude and direction of the field in which the specimen was cooled.

At this stage of the investigation, we undertook

to study the crystalline anisotropy of  $\delta(H)$ . It would have been necessary in this case to change over to flat samples, so that the field and current would have the same direction relative to the crystal axis over the entire surface of the sample. In our case the current in the sample was made to flow in circles, and we dealt always only with suitably averaged values  $\Delta_{\text{eff}}\delta$ . We therefore did not introduce in the foregoing formulas the complicating modifications necessary to allow for the anisotropy. The fact that samples 2–4, with different crystal orientations, yielded values of  $\alpha$  that were close to each other justifies this data reduction method.

We attempted also to investigate the variation of  $\delta(H)$  in a transverse field. In this case the constant and alternating fields were perpendicular to each other on the main part of the sample surface, so that the numerical coefficients in front of  $\alpha$  and  $\beta$  in (8) are changed. In the case of homogeneous crossed fields these coefficients would be unity. In our case it is necessary to allow for the fact that the constant field is different at different sections of the sample surface. Considering the sample to be a long cylinder and averaging over its perimeter, we obtain for the transverse field

$$\Delta_{\text{eff}}\delta = \delta(0) \left( \frac{1}{2} \alpha h_{\perp}^2 + \frac{3}{8} \beta h_{\perp}^4 \right), \quad (9)$$

where  $h_{\perp}$  is the ratio of the external field to the value of the field at which superconductivity begins to disappear.

The conditions for measuring  $\alpha$  were much more favorable for our samples in a transverse field than in a longitudinal field. From (8) and (9) it follows that the effect quadratic in  $h$  should be six times smaller for an isotropic sample in a transverse field than in a longitudinal one. In addition, the ends of the sample make a larger contribution to  $\Delta_{\text{eff}}\delta$  in a transverse field than in a longitudinal field, while the surface finish of the samples is undoubtedly poorer near the ends than on the cylindrical part.

The  $\Delta_{\text{eff}}(h_{\perp}^2)$  curves obtained with sample 3 disclose noticeable deviations from linearity at small values,  $h_{\perp}^2 < 0.2$ , and the slope of the curve increases with increasing field. These deviations were apparently due to loss of superconductivity of some parts of the sample surface near the ends. With increasing temperature, the deviations become less noticeable, so that a limiting value  $\alpha_{\perp} = 2 \times 10^{-2}$  could be estimated for  $T \rightarrow T_c$ . The maximum difference between the values of  $\alpha_{\perp}$ , measured at different directions of the transverse field in the horizontal plane, was 10 to 15% (the dependence of  $\alpha_{\perp}$  on the angle was sinusoidal, as

expected in a cylindrical sample, independently of the character of the true anisotropy  $\alpha$ ).

It can be concluded from all these experiments that the average value of  $\alpha$  for tin lies between  $1.4 \times 10^{-2}$  and  $2 \times 10^{-2}$  at temperatures close to  $T_c$ .

Our data give for temperatures close to  $T_c$  estimated values  $\beta = 1 \times 10^{-3}$  to  $2 \times 10^{-3}$ , which, however, must be considered only as upper limits, since the values of  $\Delta_{\text{eff}}\delta$  used for this calculation were those for large values of the external field, when loss of superconductivity on the damaged portions of the sample surface was possible.

## DISCUSSION OF RESULTS

The experimental values of  $\alpha$  obtained for  $T \rightarrow T_c$  were somewhat lower than the values calculated by the Ginzburg and Landau (GL) theory from data on the depth of penetration. According to (2) and (4),  $\alpha = 2.5 \times 10^{-2}$ , whereas the experimental values are  $\alpha = (1.4 \text{ to } 2) \times 10^{-2}$ , and the lower limit is the more reliable one, for defects in the samples cause, as has been shown, an increase in the experimental values of  $\alpha$ .

The theoretical value of  $\beta$ , calculated from (3) and (4), is  $4.5 \times 10^{-4}$ , and agrees in order of magnitude with the given experimental estimates.

It is interesting to note, that if  $\delta(0)$  is calculated from our values of  $\Delta_{\text{eff}}\delta$  and the formulas of the GL theory, the resultant value is only 10 or 15% lower than that usually assumed. A similar result can be obtained by calculating  $\delta(0)$  with the formulas of the GL theory using data on surface tension at the boundary between the superconducting and normal phases.<sup>17,18</sup> From this point of view, the discrepancies which arise in the present paper and in references 17 and 18 are of the same sign and magnitude.

In any case, the discrepancies obtained are not very large, particularly considering the inaccuracy due to neglecting the anisotropy of the effect. On the whole, our results for temperatures close to  $T_c$  should be viewed as confirming the GL theory for tin when  $T_c - T$  is on the order of  $0.1 - 0.01^\circ$ .

The results obtained should also be compared with the data on the dependence of the impedance of tin on the field, as obtained by Pippard<sup>13</sup> at 9400 Mc/sec and by Spiewak<sup>19</sup> at 1000 Mc/sec. From theoretical considerations<sup>12</sup> one could assume that all three investigations should yield like results at temperatures not too close to  $T_c$ , since the temperature used by Pippard is already quite low compared with the width  $\Delta_0$  of the energy gap for superconducting tin ( $\hbar\omega/\Delta_0 = 7 \times 10^{-2}$ ). In

fact, however, the relationships obtained from the data of these investigations differ so much in character, that a quantitative comparison becomes impossible. According to Pippard, the curve  $\alpha(T)$  has a minimum at  $3^\circ\text{K}$ , where  $\alpha$  practically vanishes (dotted curve in Fig. 6). On going to frequencies one order of magnitude lower, Spivak observed even sharper anomalies. Thus, for example, at temperatures higher than  $2.5^\circ$  the variation of the reactance with the field was non-monotonic in character, and the sign of the coefficient of  $h^2$  in the expansion in terms of the field was reversed, corresponding to a decrease in the depth of penetration after application of the field. A comparison of Pippard's and Spiewak's data with respect to the value of  $\alpha$  is possible only at temperatures lower than  $2.4^\circ\text{K}$ , where the values obtained by Spiewak are approximately twice those obtained by Pippard (according to Pippard,  $\alpha$  ranges from  $0.8 \times 10^{-2}$  to  $0.5 \times 10^{-2}$  as the temperature is increased from  $1.5$  to  $2.4^\circ\text{K}$ ). Our own measurements did not extend to this temperature region. An extrapolation of our data, which depend little on the temperature, leads to values of  $\alpha$  some two or three times greater than those obtained by Spiewak.

These discrepancies show that a further investigation of the dependence of the surface impedance of superconductors on the field at relatively low frequencies, on the order of  $10^6 - 10^9$  cps, is of undoubted interest. The results of measurements of the reactive component of the impedance at frequencies on the order of  $10^6$  cps can apparently be considered here as characterizing the static properties of superconductors.

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