

30, 834 (1959).

<sup>17</sup>For the cases in which  $T_{c3} < T_{c2}$ , we defined the reduced temperature  $t = T/T_{c3}$ . For  $T_{c2} < T_{c3}$ , we defined  $t = T/T_{c2}$ .

<sup>18</sup>S. L. Wipf, *Cryogenics* **3**, 225 (1963).

<sup>19</sup>The increase in the ratio  $H_{c3}/H_{c2}$  predicted in Refs. 1–3 was found to be zero within experimental error (1–2%). To check for this our samples were measured down to  $t = 0.5$  for the Sn-In system, and  $t = 0.3$  for the In-Bi system.

PHYSICAL REVIEW B

VOLUME 2, NUMBER 7

1 OCTOBER 1970

## Superconducting Penetration Depth of Lead<sup>†</sup>

R. F. Gasparovic\* and W. L. McLean<sup>‡</sup>

*Rutgers University, New Brunswick, New Jersey 08903*

(Received 6 April 1970)

The temperature dependence of the change in the superconducting penetration depth  $\lambda$  from its value at absolute zero has been measured in polycrystalline and single-crystal specimens of lead, using radio-frequency fields at about 80 MHz. In the polycrystalline material, it has been found that the temperature dependence is the same as that of a weak coupling superconductor with  $\Delta_T/\Delta_0$  given by the BCS theory and with an energy gap  $2\Delta_0 = 3.75kT_c$ , rather than  $2\Delta_0 = 4.3kT_c$ , the value found from tunneling measurements. Near the transition temperature  $T_c$ ,  $d\lambda/dy = 418 \pm 9 \text{ \AA}$ , where  $y = (1 - t^4)^{-1/2}$  and  $t = T/T_c$ . A full comparison with Nam's general formulation of the electromagnetic properties of superconductors (which encompasses strong coupling effects) awaits more detailed theoretical work on lead. Among the single crystals, there is a small anisotropy in the values of  $d\lambda/dy$  near  $T_c$ , and at lower temperatures a slight anisotropy in the form of the temperature dependence of  $\lambda$ . A review of all the relevant experiments on lead gives the following consistent values for the coherence length and London penetration depth of lead:  $\xi_0 = 960 \text{ \AA}$ ,  $\lambda_L(0) = 305 \text{ \AA}$ .

### I. INTRODUCTION

Lead differs from the other extensively studied superconductors in that it cannot be regarded as a close approximation to the so-called weak coupling superconductors of the Bardeen-Cooper-Schrieffer theory,<sup>1</sup> hereafter referred to as BCS superconductors. The first indications of this were deviations from the predicted temperature dependence of the critical field<sup>2</sup> and an energy gap found from infrared measurements<sup>3</sup> much larger than predicted for a superconductor with  $T_c = 7.2 \text{ K}$ . Then followed the more detailed results of tunneling experiments<sup>4</sup> supported at the same time by theoretical studies of the effects of strong coupling between the electrons and phonons in superconductors.<sup>5</sup> There still remained the question as to the effect of the strong coupling on the low-frequency electromagnetic properties, which are closely related to one of the most fundamental properties of superconductors – the Meissner effect.<sup>6</sup>

Some measurements of the penetration of radio-frequency fields by Maxfield<sup>7</sup> using the “active” resonator method<sup>8</sup> had shown an anomalous increase in the penetration as the lead was cooled through its superconducting transition, followed, at low temperatures, by the expected decrease. This anomaly was later observed by Gasparovic<sup>9</sup> using the “passive” resonator technique<sup>10</sup> which is more readily analyzed

in cases where there is doubt about what is actually being measured. At about this time, theoretical work by Scalapino and Wada<sup>11</sup> had shown that the gap function would vanish for low frequencies (of the order of the ones being used in the experiments). However, detailed calculations by Wada<sup>12</sup> showed that this had an insignificant effect on the commonly measured properties of lead. Further experimental studies of the anomalous penetration of radio-frequency fields at  $T_c$  showed that the effect was due to using too large an amplitude of oscillating field, which irreversibly forced the transition and thus produced a highly nonlinear response. Similar effects causing an apparent shift of the transition temperature had been observed by others,<sup>13</sup> but it seems that in lead greater precautions are needed than in other superconductors studied previously.

The results reported here were obtained from further measurements carried out to see if there were any real strong coupling effects on the penetration of radio-frequency fields into lead. During the course of this work, the theory of the electromagnetic properties of superconductors was extended by Nam.<sup>14</sup> His formulation is sufficiently general to include, among other things, the effects of strong coupling. This work has recently been compared with the measurements of the infrared absorption by Palmer and Tinkham<sup>15</sup> and is compared with the results of the present work later in this paper. The

extent of the comparison is limited by the present lack of calculations of the fundamental properties of lead at finite temperatures.

Calculations based on the BCS theory neglect the fact that the positive ions in the metal form a discrete lattice rather than an elastic continuum. An attempt to take into account the real lattice of lead has been made in a calculation of the superconducting energy gap by Bennett.<sup>16</sup> Limited comparisons between his results and our measurements on single crystals are made later. Because lead is a cubic crystal, the anisotropy of its penetration depth can arise only through the nonlocal nature of the relation between current density and electric field. This is not very pronounced in lead since the coherence length is not much larger than the penetration depth. Nevertheless, there is a distinct anisotropy in the penetration depth. The main reason for pursuing this aspect at some length was to ensure a further check on the possible systematic errors that can arise in experiments of the present kind. An extensive study of the gap anisotropy itself has been made by Rochlin<sup>17</sup> from tunneling experiments.

## II. THEORETICAL CONSIDERATIONS

The penetration depth of a superconductor is defined in terms of a half-space  $z > 0$  filled with the superconductor, parallel to the surface of which is a static magnetic field. The penetration depth  $\lambda$  is given by

$$\lambda = \int_0^\infty B(z) dz / B(0),$$

where  $B(z)$  is the magnetic induction at a depth  $z$  inside the superconductor. The experiments here, on the other hand, were carried out with radio-frequency fields and are more directly interpretable in terms of the inductive skin depth  $\delta_i$  and the surface resistance  $R$ . These quantities are defined as

$$\delta_i(\omega) = \text{Re} \left[ \int_0^\infty b(z) dz / b(0) \right]$$

and

$$R(\omega) = 4\pi\omega \text{Im} \left[ \int_0^\infty b(z) dz / b(0) \right],$$

where  $b(z)$  is the complex amplitude of the radio-frequency magnetic induction at a depth  $z$  and  $\omega$  is its angular frequency.  $\delta_i(\omega)$  and  $\delta_i(0) = \lambda$  are related by the Kramers-Kronig dispersion formula

$$\delta_i(\omega) - \lambda = \omega^2 \int_0^\infty d\mu R(\mu) / 2\pi^2 \mu^2 (\mu^2 - \omega^2).$$

It has been estimated<sup>18</sup> that for  $(T_c - T) > 0.04$  K, the difference between  $\delta_i$  at 80 MHz and  $\lambda$  is much less than the experimental error in the measured values of  $\delta_i$ . Since it is much easier to deal with the calculations of  $\delta_i$  in the zero-frequency limit, comparisons between our results and theory will be

confined to temperatures  $T < (T_c - 0.04$  K).

The way in which a superconductor responds to weak electromagnetic fields can be described by a linear response function  $K_{\mu\nu}(\vec{q}, \omega)$  defined by the equation

$$4\pi J_\mu(\vec{q}, \omega) = -c K_{\mu\nu}(\vec{q}, \omega) A_\nu(\vec{q}, \omega),$$

where

$$J_\mu(\vec{q}, \omega) \text{ and } A_\nu(\vec{q}, \omega)$$

are the Fourier components of wave vector  $\vec{q}$  and angular frequency  $\omega$  of the  $\mu$ th Cartesian component of the current density vector and the  $\nu$ th Cartesian component of the magnetic vector potential, respectively. It is related to the corresponding conductivity tensor by the equation

$$\sigma_{\mu\nu}(\vec{q}, \omega) = ic K_{\mu\nu}(\vec{q}, \omega) / 4\pi\omega.$$

The most general derivation of the response function for superconductors is that given by Nam,<sup>14</sup> based on the Green's-function formalism.<sup>19</sup> The results are applicable not only to weak coupling isotropic superconductors, e.g., the BCS superconductors (earlier treatments are cited in Ref. 14), but also to strong coupling superconductors, anisotropic superconductors, and superconductors with nonmagnetic or magnetic impurities.

The penetration depth  $\lambda$  can be related to the response function  $K(\vec{q}, 0)$  only when some assumption is made about the reflection of electrons at the boundary  $z = 0$  of the superconductor. Then a combination of Maxwell's equations and the equation defining  $K(\vec{q}, 0)$  can be solved for the field distribution and  $\lambda$  found from its definition. Studies of the skin effect in many normal metals by Chambers<sup>20</sup> have shown that the reflection is predominantly diffuse rather than specular. We assume that this is also true in superconducting lead, in which case the penetration depth is given by<sup>21</sup>

$$\lambda = \pi / \int_0^\infty dq \ln[1 + K(\vec{q}, 0)/q^2],$$

where

$$\vec{q} = (0, 0, q).$$

## III. EXPERIMENTAL DETAILS

### A. Method

Changes in the inductive skin depth due to changes in temperature were measured using the passive resonator technique.<sup>10</sup> The lead sample, in the form of a cylindrical rod, was surrounded by a self-resonant superconducting helix which was maintained throughout the experiment at a constant temperature to prevent effects from the changing of its own properties. The resonator was loosely coupled to a loop in the inner conductor of a transmission line - matched off resonance to a square-

law power detector – and driven by a variable-frequency constant-amplitude oscillator. From measurements of the detected power at different frequencies near resonance, the resonant frequency and bandwidth were found. The procedure was repeated at different temperatures and the changes in the inductive skin depth  $\delta_i$  and surface resistance  $R$  of the lead were obtained from the changes in resonant frequency  $f_R$  and bandwidth  $\Delta f$  using the following relations:

$$\delta_i' - \delta_i = G(f_R - f_R'), \quad R' - R = 2\pi\omega G(\Delta f' - \Delta f),$$

where  $G$  is a geometrical factor that must be found by calculation or by calibration.

### B. Apparatus

The power source was a Rohde-Schwarz – type XVD multiplier fed by a type XUA frequency synthesizer. The combination was capable of producing any frequency from 30 Hz to 300 MHz, stabilized to about 1 part in  $10^8$  over a period of a day and with greater stability during the measurement of one value of the resonant frequency. Checks were made to ensure that the dependence of the output amplitude on frequency was negligible.

Reflections in the transmission lines due to impedance mismatches were balanced out by a matching network consisting simply of a section of line of adjustable length and a variable-length tuning stub.

The detector was a 1N23 microwave diode in series with a Leeds and Northrup dc galvanometer having a current sensitivity of  $0.035 \mu\text{A}/\text{cm}$  at one meter. By comparing the actual resonance curve with a Lorentzian, power levels were chosen so that the diode operated as a square-law detector and the galvanometer deflection was thus proportional to the power transmitted to the detector.

The helix was wound from a length of superconducting wire (Nb or Nb – 25 at. % Zr) approximately equal to half the free-space wavelength of waves with frequency equal to the desired resonant frequency – in the present case, within 5 MHz of 80 MHz. The length of the helix was about 1 cm and its diameter slightly greater than the specimen diameter. It was maintained at a constant temperature by direct thermal connection to the helium bath by electrically insulating materials and a thick copper rod. That the thermal contact was adequate, even with the specimen heated above 7.2 K, was checked by replacing the specimen with one made from a normal metal.

The entire assembly, consisting of specimen, heater, helix, and thermometer, was enclosed in a copper can which was evacuated by a mechanical pump. The earth's magnetic field in the region of the specimen was compensated to a few percent us-

ing Helmholtz coils. Temperatures were determined from a previously calibrated germanium resistance thermometer using a standard three-lead arrangement with an ac bridge. Absolute temperatures below 4.2 K were accurate to  $\pm 3$  mK and above 4.2 K to  $\pm 8$  mK.

### C. Data Reduction

The equivalent circuit of the radio-frequency circuit has been analyzed elsewhere,<sup>10</sup> where the procedure for determining the resonant frequency  $f_R$  and bandwidth  $\Delta f$  is described in detail. By taking considerable care with the adjustment of the matching circuit, it was possible to make the asymmetry of the resonance curves quite small. Below the transition temperature, changes in  $f_R$  could be determined to within  $\pm 40$  Hz, which corresponds to an uncertainty in the change of inductive skin depth of approximately  $\pm 12 \text{ \AA}$ .

The uncoupled quality factor  $Q_0$  of the resonator, defined as  $Q_0 = f_R / \Delta f$ , was typically between  $6 \times 10^3$  and  $8 \times 10^3$  for  $T < T_c$ . It fell sharply, within less than a few tens of mK, to between  $1.5 \times 10^3$  and  $2 \times 10^3$  at  $T_c$  when the lead became normal.

Owing to the irregular shape of the resonator, it was not feasible to calculate the geometrical factor  $G$  relating resonant frequency changes to changes in the inductive skin depth. Instead, the large amount of experimental information on the penetration depth of tin<sup>13</sup> was used to calibrate the resonator. For each helix used in the present work,  $df_R/dy$  was measured in the range  $2.75 < y < 6.0$  for a cylinder of polycrystalline tin of approximately the same diameter as the lead specimens. Using Waldram's<sup>13</sup> value of  $d\lambda/dy = 520 \text{ \AA}$  for temperatures close to  $T_c$ , we then get  $G = 520 \text{ \AA} / |df_R/dy|$ . Corrections to allow for the small differences between the diameters of the tin and lead specimens were made by using the infinite solenoid approximation for the calculation of the self-inductance of the helix. Neglecting the error in Waldram's estimate of  $d\lambda/dy$ ,  $G$  was thus determined to within  $\pm 1.5\%$ .

The procedure used here to determine the transition temperature  $T_c$  was to choose that value which would give the best least-squares fit of the measured  $\lambda(T) - \lambda(0)$  to a linear dependence of  $y = (1 - t^4)^{-1/2}$  in the temperature range  $2.75 < y < 6.0$ . [Just below  $T_c$ , the relation between current density and vector potential is local.<sup>19</sup> If we assume the Ginzburg-Landau theory<sup>19</sup> to be applicable,<sup>22</sup> then  $\lambda(T) \propto (1 - t)^{-1/2}$  for  $t = T/T_c \sim 1$ , but below the critical fluctuation region. For  $t \sim 1$ ,  $y \sim \frac{1}{2}(1 - t)^{-1/2}$ .] The value of  $T_c$  obtained in this way was always within a few mK of the lowest temperature at which the bandwidth of the resonance curve was indistinguishable from its value in the normal state. This

appeared to be a more precise way of defining  $T_c$  than from measurements of the bandwidth itself. Varying the choice of  $T_c$  by  $\pm 0.001$  K changes the value near  $T_c$  of  $d\lambda/dy$  by less than 1.5%.

#### D. Specimen Preparation

The lead specimens were prepared from 99.9999%-purity starting material obtained from Cominco American, Inc. (Spokane, Wash.) in the form of lead shot. After etching to remove any oxide layer, the shot was melted in a graphite mold in a vacuum to form a cylindrical rod and slowly cooled to room temperature. Polycrystalline specimens of about 2.5 in. in length were carefully machined to the desired diameter (approximately 0.300 in.), etched, and then annealed just below the melting point for approximately 100 h. Some single crystals were grown by just slowly cooling the melt in an appropriate temperature gradient, whereas others were grown from short small-diameter seeds inserted into the bottom of the mold. To avoid introducing irreparable damage, the single crystals were not machined.

As surface roughness can markedly alter the experimental results, considerable effort was expended in developing suitable polished surfaces. Several electropolishing solutions described by Tegart<sup>23</sup> were tried with no consistently reproducible results. Satisfactory surfaces were prepared, however, using the following solutions: an etching solution of 16 parts water, 4 parts acetic acid, and 3 parts nitric acid; a chemical polish of 4 parts acetic acid and 1 part 30% solution of hydrogen peroxide. With freshly prepared solutions, the specimen was alternately etched, washed in water, and polished to the desired diameter. After rinsing in water, the specimen was sprayed with acetone. Upon drying, this left a protective film which prevented rapid surface oxidation. Surfaces prepared in this manner were free from pits, uniformly polished, and appeared highly reflecting when observed under a moderate-power microscope.

### IV. EXPERIMENTAL RESULTS AND ANALYSIS

#### A. Polycrystals

In comparing the experimental results with theory, we assume that it should be possible to describe the polycrystalline specimens by the theory for an isotropic superconductor, the parameters of which are obtained from other experimental knowledge by the method described in the Appendix.

The circles in Fig. 1 show the experimental results for a polycrystalline specimen, whereas the solid curve was calculated using the weak coupling limit of Nam's<sup>14</sup> theory together with the assumptions listed below. Because only changes in  $\lambda$  were

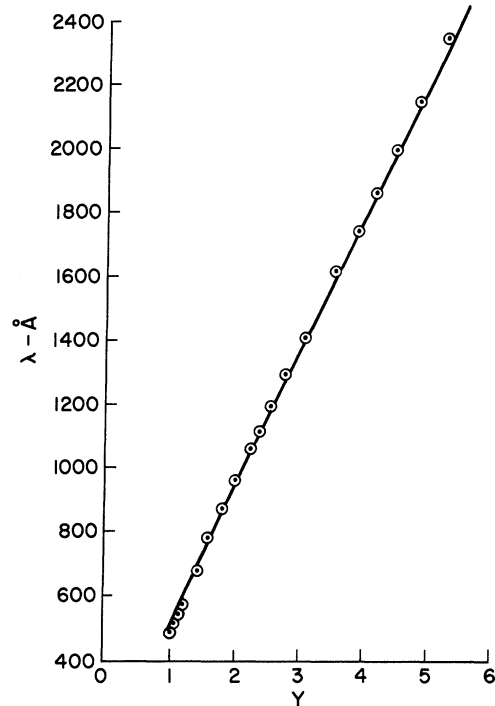


FIG. 1. Experimentally measured penetration depth as a function of  $y = (1 - t^4)^{-1/2}$  for a polycrystalline specimen. The solid line was calculated using Nam's theory (Ref. 14) and the superconducting parameters given in the text. Similar experimental results, within the errors quoted in the text, were obtained with other polycrystalline specimens of high surface quality.

measured, the arbitrary additive constant has been chosen to make the measurements and theory agree on the graph at the point  $y = 2.75$ . The slope found experimentally was  $d\lambda/dy = (418 \pm 9)$  Å for  $2.75 < y < 6.0$ . The assumptions made in obtaining the calculated curve were as follows:  $\xi_0 = 960$  Å and  $\xi_0/\lambda_L(0) = 3.12$  (see Appendix); that the electron mean free path in the normal state was infinite; and that the temperature dependence of  $\Delta_T/\Delta_0$  was the same as predicted by BCS (as has actually been found to be the case from tunneling experiments<sup>24</sup>). The value of  $\Delta_0$  was then chosen to make the calculated slope  $d\lambda/dy$  equal to the measured slope in the range  $2.75 < y < 6.0$ . This procedure yielded  $2\Delta_0 = 3.75kT_c$  rather than  $4.3kT_c$ , the value found from tunneling measurements. The interpretation of these results is discussed in Sec. V.

As far as the behavior of  $d\lambda/dy$  at temperatures below  $y \sim 2.75$  is concerned, it can be seen from Fig. 1 that there is a marked increase in the slope  $d\lambda/dy$  as  $y \rightarrow 1$ , which happens to agree quite closely with the increase in the slope predicted by the BCS theory. Because of anticipated strong coupling effects, this agreement is more likely to be acciden-

tal than of fundamental significance. It is worth recalling that, in most other superconductors previously studied, the slope increases less rapidly than predicted by BCS – presumably, as pointed out by Waldram,<sup>13</sup> because their energy gaps do not vary with temperature in exactly the same way as a BCS superconductor.

### B. Single Crystals

The specimens used in these experiments were all in the form of long circular cylinders. Figure 2 is a stereographic projection showing for each specimen the orientation of the cylinder axis relative to the cubic crystal axes. The [100] axis is normal to the plane of the stereogram, and [010] is parallel to the line joining the points marked [100] and [110]. The number beside each point identifies the specimen.

For each crystal, the slope  $d\lambda/dy$  was determined from a least-squares fit to the measurements in the range  $2.75 < y < 6.0$ . The results are given in Table I. It should be noted that the results obtained from crystals 11 and 13, which differed in orientation by 1 degree, do agree within the estimated experimental errors, indicating that small differences in surface quality produce no significant effect. Shown also in Table I are values of  $2\Delta_0/kT_c$  deduced from the weak coupling theory.

From Table I we see that the maximum anisotropy observed in  $d\lambda/dy$  was approximately 10%. To account properly for these results – including the strong coupling effects as well as the anisotropy of the electron distribution in reciprocal space and the anisotropy of the phonons – would be a formidable task which there is little hope of carrying out in the near future, if only because of the lack of detailed theoretical results referred to in the Introduction. A further complication arises because the nearly circular current-flow paths around the spec-

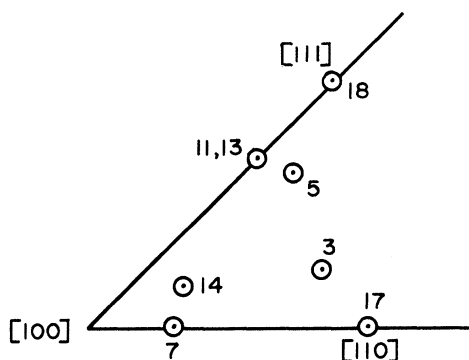


FIG. 2. Stereographic projection showing the orientation of the axis of each specimen relative to the cubic crystal axes. The numbers identify the specimen.

TABLE I. Slope  $d\lambda/dy$  determined from a least-squares fit to the measurements in the range  $2.75 < y < 6.0$ .

Specimen	$d\lambda/dy$ (in Å)	$2\Delta_0/kT_c^a$
3	$425 \pm 9$	$3.70 \pm 0.08$
5	$438 \pm 6$	$3.58 \pm 0.05$
7	$399 \pm 17$	$3.92 \pm 0.17$
8 <sup>b</sup>	$418 \pm 9$	$3.75 \pm 0.08$
11	$425 \pm 8$	$3.70 \pm 0.07$
13	$420 \pm 8$	$3.73 \pm 0.07$
14	$411 \pm 7$	$3.81 \pm 0.06$
17 [110]	$430 \pm 10$	$3.66 \pm 0.09$
18 [111]	$442 \pm 7$	$3.56 \pm 0.06$

<sup>a</sup>Deduced from second column using weak coupling theory.

<sup>b</sup>Polycrystalline.

imens reduce the anisotropy in the quantity actually measured below what would be expected for plane surfaces. All of these effects have been discussed in some detail by Gasparovic.<sup>18</sup> We note here only that there is a correlation between the relative anisotropy of the values of the energy gap found by using the weak coupling theory (in the same way as for the polycrystalline specimen) and the relative anisotropy of the energy gap calculated by Bennett.<sup>16</sup> In particular, the greatest observed deviation from the mean "gap" is about 4.8%, whereas Bennett found it to be 5.9%. Also, for current flow normal to [100], the trend of the measurements indicates that the penetration depth is less than in the polycrystalline specimen, in agreement with deductions from Bennett's results.

In addition to the anisotropy of the values of  $d\lambda/dy$  at large values of  $y$ , we have also observed a slight but definite anisotropy in the temperature dependence of the penetration depth. Figure 3 is a plot of  $\Delta\lambda = \lambda(T) - \lambda(0)$  versus  $y$  for the [111] and [110] crystals. The results for the other crystals lie in between these two curves.<sup>25</sup> Although the weak coupling formalism has been used extensively above as a way of presenting the results, it does not seem useful to try to interpret the anisotropy of the temperature dependence of  $\lambda$  in lead by the technique used by Waldram<sup>13</sup> for tin – i. e., in terms of the temperature dependence of the gap being different from that predicted by the BCS theory.

### C. London Penetration Depth $\lambda_L(0)$

In view of the general interest in the value of this parameter, we give here as accurate an estimate of it as we can from the present results. Because of the strong coupling effects, there is no simple formula for inverting the experimentally measured quantities to obtain  $\lambda_L(0)$ , the London penetration depth at absolute zero. However, it is possible to place an upper limit on the possible value of  $\lambda_L(0)$

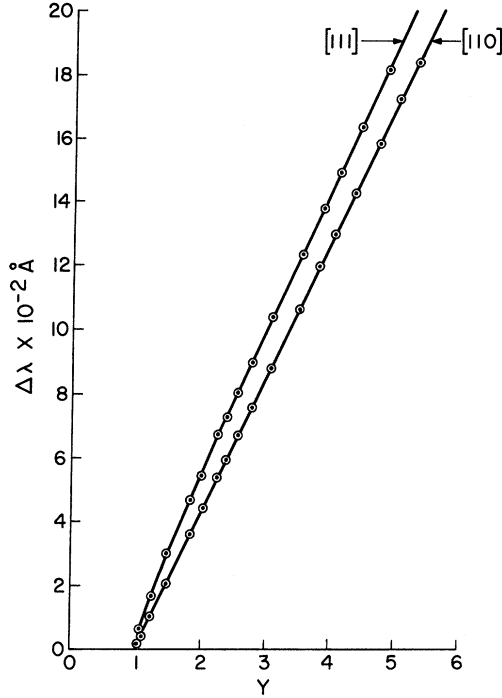


FIG. 3. The measured change in penetration depth as a function of  $\gamma$  for a [111] and a [110] crystal. The results for the other crystals lay in between these two curves (Ref. 25).

in the following way: In a weak coupling superconductor  $\lambda(T) \rightarrow \lambda_L(T)$  as  $T \rightarrow T_c$ . The comparison of the measured  $d\lambda/dy$  and of Palmer and Tinkham's<sup>15</sup> results with the weak coupling theory indicates that the effect of the strong coupling is to increase the penetration depth above its weak coupling value; thus, in the range  $2.75 \leq y \leq 6.00$ ,  $d\lambda_L/dy \leq d\lambda/dy$ . The weak coupling form of Nam's theory together with the BCS values of  $\Delta_T/\Delta_0$  and  $2\Delta_0 = 3.75kT_c$  give  $d\lambda_L/dy = 1.34\lambda_L(0)$ . Thus, we estimate  $\lambda_L(0) \leq 312 \text{ \AA}$ .

It is difficult to compare the only earlier measurement of the penetration depth in lead with the present results – or even with existing theory. From magnetization measurements of thin films, Lock<sup>26</sup> estimated  $[\lambda(4.24 \text{ K})/y(4.24 \text{ K})] = 390 \pm 30 \text{ \AA}$ . However, the method of interpretation assumed that the fields in the films obeyed the London equation. If this were true because of short mean free path effects, the result would need to be corrected to give the infinite mean free path penetration depth. Comparison with our results could still not be made directly since in the present experiment only changes in the penetration depth and not its absolute value were measured. Also, for the reasons given earlier, it is not feasible at the present time to compute the penetration depth at 4.24 K from Nam's theory.

#### D. Normal-State Measurements

By measuring the difference between the normal-state inductive skin depth and the penetration depth at  $T = 0$ , as well as the corresponding change in bandwidth of the resonance curve, the ratio of the imaginary to the real part of the normal-state surface impedance  $X_n/R_n$  can be found. For the single-crystal specimens, the average  $X_n/R_n = 1.50 \pm 0.04$ . The anomalous skin effect theory gives  $X_n/R_n = 3^{1/2} = 1.732$  in the limit of infinite mean free path. From resistivity measurements on two crystals, the mean free path at 7 K was estimated to be approximately  $4.6 \times 10^{-4} \text{ cm}$ . The anomalous skin effect formula, corrected for finite mean free path,<sup>27</sup> then gives  $X_n/R_n = 1.48$ , in reasonable agreement with the average of the various crystals.

The mean free path dependence of the penetration depth is governed by the size of the quantity  $\pi\xi_0/l$ . On evaluating Nam's expressions for  $\lambda$  as a function of  $l$ , the infinite mean free path limit is effectively achieved if  $\pi\xi_0/l < 0.1$ . For our specimens at 7 K, we estimate  $\pi\xi_0/l \approx 0.07$  so that they can reasonably be compared with the infinite mean free path calculations of  $\lambda$ .

#### V. DISCUSSION OF THE STRONG COUPLING EFFECTS

In Sec. IV A it was pointed out that the temperature dependence of the penetration depth was found to be the same, within experimental error, as predicted in the BCS theory but with an energy gap less than the value of  $2\Delta_0 = 4.3kT_c$  found from tunneling measurements, namely,  $2\Delta_0 = 3.75kT_c$ . Similar behavior has been found in the microwave surface resistance of lead by Turneaure<sup>28</sup> [with  $2\Delta_0 = (4.02 \pm 0.06)kT_c$ ] and by Hahn *et al.*<sup>29</sup> (with  $2\Delta_0 = 3.94kT_c$ ), and also in the ultrasonic attenuation in lead<sup>30</sup> (with results ranging from  $2\Delta_0 = 3.5kT_c$  to  $2\Delta_0 = 4.0kT_c$ , the lower values being the most frequently found).

The microscopic properties of lead at absolute zero have been worked out in sufficient detail to enable a full comparison of Nam's theory to be made with the imaginary part of the conductivity deduced from Palmer and Tinkham's<sup>15</sup> measurements of the far-infrared absorption in lead films at  $T \ll T_c$ . However, it is worth noting that, within the experimental error, equally close agreement with the infrared results is obtained by using the weak coupling theory with  $2\Delta_0 = 3.75kT_c$ , the value found in Sec. IV A.

The above correspondences between the measured properties of lead and the weak coupling theory are probably accidental rather than of fundamental significance, although it is interesting that the different phenomena do involve the response function  $K(\vec{q}, \omega)$  evaluated at quite different wave vectors and frequencies. A proper comparison with the general formulation of the electromagnetic properties of

superconductors by Nam will be possible when more details of the theoretical work on lead at finite temperatures become available.

#### ACKNOWLEDGMENTS

We would like to acknowledge many useful discussions with Dr. S. B. Nam and are grateful for his aid in the numerical computations. We also thank A. Gynn for his assistance in the preparation and orientation of the crystals.

#### APPENDIX: PARAMETERS OF POLYCRYSTALLINE LEAD

The parameters required for the application of the isotropic weak coupling theory are the London penetration depth at absolute zero  $\lambda_L(0)$  and the coherence length  $\xi_0$ .

The first of these quantities, for a cubic crystal, can be expressed in the form<sup>31</sup>

$$\lambda_L(0) = 3\hbar \langle v \rangle_F / \langle 1/v \rangle_F^{1/2} / e k S,$$

where  $\langle v \rangle_F$  is the average of the velocity over the Fermi surface,  $S$  is the area in one Brillouin zone of all sheets of the Fermi surface excepting those parts that are in contact with the zone boundary, and  $\gamma$  is the coefficient of the temperature in the low-temperature electronic specific heat. It is usually assumed that  $\langle 1/v \rangle_F = 1/\langle v \rangle_F$ . Although this result was derived<sup>31</sup> using the independent electron model, it should still remain valid in the quasiparticle description which includes both the Coulomb repulsion between the electrons and also the electron-phonon interaction. It has been shown by Heine *et al.*<sup>32</sup> that, provided the Coulomb repulsion is included in the band-structure calculation, the effect of then introducing the electron-phonon interaction is to leave the Fermi surface unchanged (in shape and size) but to change ("renormalize") the velocity on the Fermi surface. Owing to the anisotropy of the electron-phonon interaction itself,<sup>33</sup> the anisotropy of the renormalized Fermi velocity will not, in general, be the same as the unrenormalized Fermi velocity; but without a detailed calculation it is not possible to tell whether the difference between  $\langle 1/v \rangle_F$  and  $1/\langle v \rangle_F$  is made greater or less by renormalization.

There have been many estimates of  $S$ . From anomalous skin effect measurements, Chambers<sup>20</sup> deduced  $S/S_0 = 0.46$ , where  $S_0$  is the free-electron

value for an electron density equal to the valence electron density of lead, but he noted that this was probably an underestimate because of the effects of surface roughness. Aubrey<sup>34</sup> found from cyclotron resonance experiments that  $S/S_0 = 0.55$ . Anderson and Gold<sup>35</sup> estimated  $S/S_0 = 0.59$  from de Haas-van Alphen effect experiments, but this was later corrected by Ashcroft<sup>36</sup> to  $S/S_0 = 0.70$ . Observations of the Kohn anomalies in the phonon dispersion curves, by Stedman *et al.*,<sup>37</sup> also gave  $S/S_0 = 0.70$ . This value appears to be the best available estimate and is the value that has been used in the analysis of many experiments on lead.<sup>27,28,38,39</sup> The ratio  $\gamma/\gamma_0$ , where  $\gamma_0$  is the free-electron value, has been found from specific-heat measurements<sup>40</sup> to be 2.1.

The other parameter of interest in the isotropic weak coupling case is the coherence length  $\xi_0 = \hbar v_F / \pi \Delta_0$  (see Sec. II). In an anisotropic superconductor, both  $v_F$  and  $\Delta_0$  will vary over the Fermi surface. We again assume that the many-body effects can be included by the renormalization of  $v_F$  and merely use for  $v_F$  in this formula the reciprocal of the average of the reciprocal of the Fermi velocity obtained from the relation  $\gamma = S v_F \gamma_0 / S_0 v_F$ , where  $v_{F0}$  is the free-electron Fermi velocity. Assuming four valence electrons per lead atom,  $v_{F0} = 1.82 \times 10^8$  cm/sec. Thus, using  $\gamma/\gamma_0 = 2.1$  and  $S/S_0 = 0.70$ , we get  $v_F = 0.61 \times 10^8$  cm/sec.

A review of estimates of  $\Delta_0$  from infrared and tunneling experiments is given by Palmer and Tinkham.<sup>15</sup> The generally accepted value is  $2\Delta_0 = 4.3 k T_c$ .<sup>24</sup>

The commonly quoted value of  $\xi_0$  for lead<sup>41</sup> is 830 Å which is arrived at by using Aubrey's estimate of  $S/S_0$  to obtain  $v_F$  and the value  $2\Delta_0 = 4.1 k T_c$ , as found from early infrared measurements.<sup>3</sup> However, using  $v_F = 0.61 \times 10^8$  cm/sec and  $2\Delta_0 = 4.3 k T_c$  we get  $\xi_0 = 960$  Å. Again using  $S/S_0 = 0.70$  and the measured  $\gamma = 2.08 \gamma_0$ , we calculate from the formula above,  $\lambda_L(0) = 305$  Å.

The Ginzburg-Landau parameter at  $T_c$  in the infinite mean free path limit for weak coupling superconductors is given by  $\kappa = 0.96 \lambda_L(0) \Delta_0^{\text{BCS}} / \xi_0 \Delta_0$ , where  $2\Delta_0^{\text{BCS}} = 3.5 k T_c$ . We thus get  $\kappa = 0.25$ , which should be compared with the value of  $\kappa = 0.24$  found from the experiments of Smith and Cardona.<sup>38</sup>

A similar self-consistent analysis of the parameters of aluminum, indium, niobium, and tin has been carried out by Gasparovic.<sup>18</sup>

<sup>†</sup>Supported by the National Science Foundation and the Rutgers Research Council.

\*Present address: RCA Advanced Technology Laboratories, Camden, N. J. 08102.

<sup>‡</sup>Part of this work was completed in the Department of Physics, University of California, Berkeley, with the support of a Rutgers University Research Council Faculty

Fellowship.

<sup>1</sup>J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

<sup>2</sup>D. K. Finnemore, E. E. Mapother, and R. W. Shaw, Phys. Rev. **118**, 127 (1960).

<sup>3</sup>P. L. Richards and M. Tinkham, Phys. Rev. **119**, 575 (1960).

- <sup>4</sup>J. M. Rowell and L. Kopf, Phys. Rev. **137**, A907 (1965).
- <sup>5</sup>D. J. Scalapino, J. R. Schrieffer, and J. W. Wilkins, Phys. Rev. **148**, 263 (1966).
- <sup>6</sup>W. Meissner and R. Ochsenfeld, Naturwiss. **21**, 787 (1933).
- <sup>7</sup>B. W. Maxfield (unpublished).
- <sup>8</sup>A. L. Schawlow and G. E. Devlin, Phys. Rev. **113**, 120 (1959).
- <sup>9</sup>R. F. Gasparovic (unpublished).
- <sup>10</sup>W. L. McLean, Proc. Phys. Soc. (London) **79**, 572 (1962).
- <sup>11</sup>D. J. Scalapino, Y. Wada, and J. C. Swihart, Bull. Am. Phys. Soc. **9**, 267 (1964).
- <sup>12</sup>Y. Wada (unpublished).
- <sup>13</sup>See J. R. Waldram, Advan. Phys. **13**, 1 (1964), and references therein.
- <sup>14</sup>S. B. Nam, Phys. Rev. **156**, 470 (1967); **156**, 487 (1967).
- <sup>15</sup>L. H. Palmer and M. Tinkham, Phys. Rev. **165**, 588 (1968).
- <sup>16</sup>A. J. Bennett, Phys. Rev. **140**, A1902 (1965).
- <sup>17</sup>G. I. Rochlin, Phys. Rev. **153**, 513 (1967).
- <sup>18</sup>R. F. Gasparovic, Ph.D. thesis, Rutgers University, 1969 (unpublished).
- <sup>19</sup>J. R. Schrieffer, *Theory of Superconductivity* (Benjamin, New York, 1964).
- <sup>20</sup>R. G. Chambers, Proc. Roy. Soc. (London) **A215**, 481 (1952).
- <sup>21</sup>E. A. Shapoval, Zh. Eksperim. i Teor. Fiz. **47**, 1007 (1964) [Soviet Phys. JETP **20**, 675 (1965)].
- <sup>22</sup>A derivation from microscopic theory of the Ginzburg-Landau equations for strong coupling superconductors has been given by N. Menyhård, Nuovo Cimento **44B**, 213 (1966); see also, G. Eilenberger and V. Ambegaokar, Phys. Rev. **158**, 332 (1967); E. D. Yorke and A. Bardasis, *ibid.* **159**, 344 (1967), whose results are partially confirmed experimentally by Ref. 38 below.
- <sup>23</sup>W. J. McG. Tegart, *Electrolytic and Chemical Polishing of Metals* (Pergamon, London, 1956).
- <sup>24</sup>R. F. Gasparovic, B. N. Taylor, and R. E. Eck, Solid State Commun. **4**, 59 (1966).
- <sup>25</sup>Detailed plots of  $\lambda(T) - \lambda(0)$  as functions of  $y(T)$  can be obtained from one of the authors (RFG).
- <sup>26</sup>J. M. Lock, Proc. Roy. Soc. (London) **A208**, 391 (1951).
- <sup>27</sup>K. R. Lyall and J. F. Cochran, Phys. Rev. **159**, 517 (1967).
- <sup>28</sup>J. P. Turneaure, Stanford University, High Energy Physics Laboratory HEPL Report No. 507, 1967 (unpublished).
- <sup>29</sup>H. Hahn, H. J. Halama, and E. H. Foster, J. Appl. Phys. **39**, 2606 (1968).
- <sup>30</sup>B. C. Deaton, Phys. Rev. **177**, 689 (1969).
- <sup>31</sup>T. E. Faber and A. B. Pippard, Proc. Roy. Soc. (London) **A231**, 336 (1955).
- <sup>32</sup>V. Heine, P. Nozières, and J. W. Wilkins, Phil. Mag. **13**, 741 (1966).
- <sup>33</sup>See N. W. Ashcroft and W. E. Lawrence, Phys. Rev. **175**, 938 (1968), and references therein.
- <sup>34</sup>J. E. Aubrey, Phil. Mag. **5**, 1001 (1960).
- <sup>35</sup>J. R. Anderson and A. V. Gold, Phys. Rev. **139**, A1459 (1965).
- <sup>36</sup>N. W. Ashcroft, Ph.D. thesis, University of Cambridge, 1964 (unpublished).
- <sup>37</sup>R. Stedman, L. Almquist, G. Nilsson, and G. Rannio, Phys. Rev. **163**, 567 (1967).
- <sup>38</sup>F. W. Smith and M. Cardona, Solid State Commun. **6**, 37 (1968).
- <sup>39</sup>G. Grimvall, Physik Kondensierten Materie **8**, 202 (1968).
- <sup>40</sup>B. J. C. van der Hoeven and P. H. Keesom, Phys. Rev. **137**, A103 (1965).
- <sup>41</sup>J. Bardeen and J. R. Schrieffer, in *Progress in Low-Temperature Physics*, edited by C. J. Gorter (Interscience, New York, 1961), Vol. 3.

## Type-I and Type-II Superconductivity in Wide Josephson Junctions

Klaus Schwidtal

*Institute for Exploratory Research, U. S. Army Electronics Command, Fort Monmouth, New Jersey 07703*

(Received 7 May 1970)

The magnetic field dependence of the maximum dc Josephson current  $I_{\max}$  of wide Pb-PbO<sub>x</sub>-Pb Josephson junctions, for which the junction length  $L$  was about 10 times the Josephson penetration depth  $\lambda_J$ , has been measured. The experimental data are in excellent agreement with a theoretical prediction by Owen and Scalapino, who have numerically calculated  $I_{\max}$  versus  $H_e$  for a Josephson junction with  $L = 10\lambda_J$ , and who have also shown that the Meissner region and the vortex structure are reflected in the  $I_{\max}$ -versus- $H_e$  curve in a very characteristic way. It is shown that the boundary conditions of Owen and Scalapino's model can be most conveniently fulfilled with a cross-type junction configuration, and that the PbO<sub>x</sub> tunneling barrier layers were fairly uniform.

### I. INTRODUCTION

Wide Josephson junctions, i.e., junctions whose dimension  $L$  perpendicular to an externally applied

magnetic field  $H_e$  is large with respect to the Josephson penetration depth  $\lambda_J$ , behave like an extreme type-II superconductor; they exhibit a Meissner effect in weak magnetic fields, and vortex