# Pressure dependence of the critical magnetic field of superconducting lead*

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The pressure dependence of the critical magnetic field of lead has been measured in the range 0.3–4.2 K. The measurements were done using hydrostatic pressures up to 3 kbar. The pressure derivatives of $H_0$ and $\gamma^*$ are reported, where $H_0$ is the extrapolated critical magnetic field at 0 K and $\gamma^* = \gamma/V$, where $\gamma$ is the temperature coefficient of the normal electronic specific heat and $V$ is the molar volume. $\partial \ln H_0/\partial \ln V = 4.77 \pm 0.12$ and $\partial \ln \gamma^*/\partial \ln V = 3.6 \pm 0.4$. The shape of the experimental deviation function $D(t) = h - (1-t^2)$, where $h = H/H_0$ and $t = T/T_c$, is shown to change under pressure, in agreement with the strong-coupling theory of superconducting lead under pressure.

## I. INTRODUCTION

Critical-field measurements can be extrapolated to 0 K to determine the values of $H_0$ and $\gamma^*$, where $\gamma^* = \gamma/V$. ($\gamma$ is the temperature coefficient of normal electronic specific heat and $V$ is the molar volume.) The value of $\gamma^*$ obtained in this way is dependent on the temperature range of the extrapolation, which is determined by the experimental conditions. Accurate extrapolation requires data in the range below $0.25T_c$, where $T_c$ is the critical temperature. Previous experimental data for Pb (Refs. 1 and 2) were taken at temperatures above $0.20T_c$ and pressures up to 0.7 kbar. Therefore, the method of direct extrapolation at low temperatures was not applicable, and the similarity principle was used for analyzing the results. This semiempirical principle assumes that $H_c(P, T) = H_0(P)f(T)$, where $H_c(P, T)$ is the general shape of the critical field as a function of temperature and pressure, and $H_0(P)$ and $f(T)$ are experimental one-parameter functions.

More recent theoretical derivations have shown that the similarity principle is not valid for strong-coupling superconductors, especially for Pb.[3] Furthermore, the result obtained for the electronic Grüneisen parameter[1] $\partial \ln \gamma^*/\partial \ln V = 4.9$ was in disagreement with the value 0.7 deduced from thermal-expansion experiments.[4]

The fine details of the critical-field curve are revealed by the deviation function $D(t) \equiv h - (1 - t^2)$ where $h = H_c/H_0$ and $t = T/T_c$ are the reduced critical field and temperature, respectively. The shape of $D(t)$ is a function of the electron-phonon interaction strength. As pressure is applied on a strong-coupling superconductor, the strength of the coupling usually decreases toward the BCS value and the shape of $D(t)$ follows accordingly, i.e., the $D(t)$ curve is displaced toward negative values.[5] As the shape of $D(t)$ changes, the biggest absolute decrease is expected near the peak (for Pb the peak is at $t^2 \cong 0.40$). A quantitative calculation[3] of this change was done for Pb in the very-low-temperature limit ($t^2 \ll 1$ or $t < 0.1$).

The present experiment was designed to measure the parameters $d \ln H_0/d \ln V$ and $d \ln \gamma^*/d \ln V$, and to find out directly whether the shape of $D(t)$ changes under pressure. A $He^3$ refrigerator enabled us to measure down to 0.3 K or $0.05T_c$ while the pressure used was 3 kbar. We have been able to estimate the volume derivatives of $H_0$ and $\gamma^*$ by direct extrapolation without using the similarity principle assumption. The results will be compared to the theoretical[6,7] and experimental picture of strong-coupling Pb which includes some recent tunneling[8–11] and volume-change[12] measurements.

## II. EXPERIMENTAL TECHNIQUE

### A. Apparatus

The apparatus used in the present work incorporates a hydrostatic high-pressure system and a standard $He^3$ refrigerator. The pressure transmitting medium was solid $He^4$ which is believed to be the best way to achieve true hydrostatic pressure.[13]

The pressure cell is shown in Fig. 1. The heat-treated Be-Cu cell (a cylinder $\frac{3}{8}$-in. o.d. and $\frac{1}{8}$-in. i.d.) had two compartments: one for an unpressurized reference sample and one connected to the external pressure system by means of capillaries ($\frac{1}{32}$-in. o.d. and 0.007-in. i.d. made of stainless steel). The pressure in the cell was measured to better than 2% accuracy by commerical strain gauges.[14–15] A bridge arrangement using four strain gauges eliminated any temperature dependence and increased the sensitivity. The absolute calibration of the pressure was done using the pressure dependence of $T_c$ of In. This quantity had a very well accepted value.[14,16] In the actual measurement a lock-in amplifier was used to drive and measure the bridge, thus achieving a high sensitivity.

The $He^3$ refrigerator unit was of standard type.[5] The Be-Cu pressure cell was attached to the bottom of the $He^3$ chamber by a thermally contracted

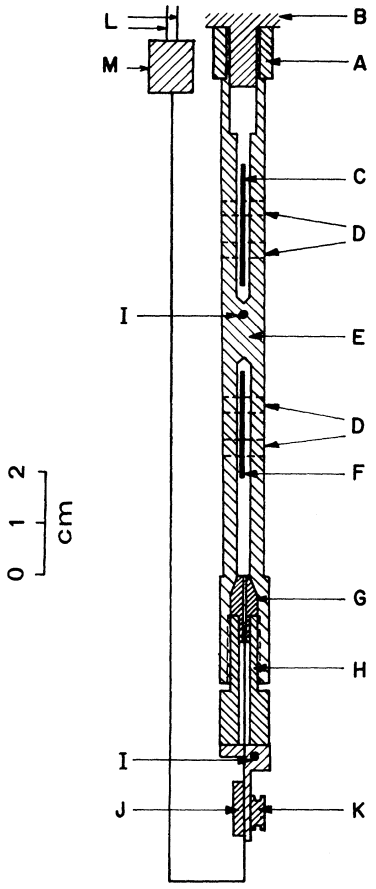


FIG. 1. The experimental Be-Cu high-pressure cell. A, Nylon clamping ring. B, Bottom part of a $He^3$ refrigerator. C, Reference sample (sample holder not shown). D, Strain gauges. E, Be-Cu cylinder. F, Pressurized sample. G and H, High-pressure seal. I, Resistance thermometers for the temperature stabilizer. J, Clamping plate for the high pressure capillaries. K, Heater. L, Capillaries. M, Joint for capillaries.

nylon ring (A in Fig. 1) thus providing excellent contact.[17] The temperature was controlled by an electronic regulator[18] having an assembly of ten parallel $\frac{1}{2}$-W, 470-Ω Speer carbon resistors as a temperature sensor.

The highly homogeneous magnetic field applied to the samples was produced by an aluminum-foil nitrogen-cooled solenoid as described by Gubser *et al.*[19] The isothermal magnetization measurements were done by a pick-up coil mounted on a piezoelectric vibrator.[20] The procedure was similar to that used by Gubser *et al.*[19] except that only one coil was used. To assure the same measuring conditions for the two samples, the solenoid and the pick-up coil were displaced vertically for each measurement thus avoiding systematical errors in the magnetization measurement.

### B. Samples

The cylindrical polycrystalline samples (1×0.045-in. diam) were made of 99.999% pure lead obtained from the American Smelting and Refining Company. The unprocessed lead rods were swaged and then chemically polished to the desired diameter. The samples were annealed for 48 h at 322°C in $\frac{1}{3}$ atm of pure He.

The resulting samples possessed a highly hysteretic magnetization curve.[21] This hysteresis was explained[22,23] by the formation of a superconducting surface sheath that is metastable above $H_c$ and shields the applied external field. The experimental solution of this problem is to fix the order parameter at zero on the surface by coating the sample with a thin layer of a magnetic material, like Cr, Ni, or Mn.[19] For our Pb samples we used a ≈ 200-Å uniform layer of Mn, with an additional layer of SiO to prevent oxidation. The coated samples showed very small hysteresis (up to 0.2 G at fields of 800 G).

### C. Procedure

Two identical samples were placed in the two compartments of the Be-Cu pressure cell (Fig. 1). The capillaries and the cell, while thermally isolated from the external $He^4$ bath, were heated to several degrees above the melting temperature of $He^4$ at the desired pressure. By circulating $He^3$ through the $He^3$ chamber a slow, almost constant pressure freezing occurred in the cell. The *in situ* measurement by the strain gauges gave the value of the pressure within the cell at the end of the freezing procedure.

A zero pressure run over the entire temperature range from 0.3 to 4.2 K showed that the heat conduction along the pressure cell was adequate for isothermal conditions: the temperature difference between the two samples was smaller than 0.5 mK.

Each transition was measured twice: from the superconducting to the normal state and vice versa. To eliminate the problem of trapped flux the external field was reduced to zero before each measurement. The field sweep speed was determined by the reversibility requirement during the transition.[19] The very small hysteresis observed for the coated samples at low temperatures was corrected by assuming that the thermodynamical critical field is the middle of the hysteresis loop.[21]

Only the highest pressure (3 kbar at 4.2 K) measurement will be reported, although a 2-kbar run was done to check the reproducibility of the data.

## III. RESULTS

### A. Data analysis

The critical field $H_c = H_c(P, T)$ is in general a function of the pressure and temperature. At suf-

TABLE I. Isothermal observations of $H_c(P, T)$ at $P = 0$ and $P = 2960$ atm.

| $H_c(P=0, T)$ (G) | $H_c(P=2960$ atm, $T)$ (G) | $T$(K) | $H_c(P=0, T)$ (G) | $H_c(P=960$ atm, $T)$ (G) | $T$(K) |
|---|---|---|---|---|---|
| 546.92 | 520.91 | 4.189 | 719.64 | 695.54 | 2.446 |
| 553.31 | 527.39 | 4.140 | 728.58 | 704.40 | 2.314 |
| 569.13 | 543.20 | 4.015 | 740.37 | 716.41 | 2.125 |
| 595.57 | 569.95 | 3.793 | 743.37 | 719.57 | 2.073 |
| 610.08 | 584.59 | 3.665 | 744.17 | 720.29 | 2.059 |
| 619.38 | 594.02 | 3.580 | 749.04 | 725.14 | 1.973 |
| 635.84 | 610.67 | 3.423 | 758.58 | 734.84 | 1.791 |
| 641.83 | 616.81 | 3.364 | 759.31 | 735.60 | 1.774 |
| 652.29 | 627.30 | 3.258 | 764.89 | 741.04 | 1.655 |
| 657.35 | 632.34 | 3.205 | 768.17 | 744.46 | 1.581 |
| 668.55 | 643.70 | 3.084 | 774.40 | 750.85 | 1.428 |
| 685.12 | 660.52 | 2.895 | 785.47 | 761.86 | 1.102 |
| 695.32 | 670.92 | 2.771 | 786.19 | 762.57 | 1.078 |
| 699.49 | 675.03 | 2.718 | 797.36 | 773.89 | 0.562 |
| 705.94 | 681.73 | 2.635 | 798.13 | 774.65 | 0.507 |
| 708.97 | 684.66 | 2.594 | 798.97 | 775.55 | 0.439 |
| 710.90 | 686.61 | 2.568 | 799.95 | 776.43 | 0.343 |
| 715.23 | 691.16 | 2.508 | 800.30 | 776.88 | 0.302 |

ficiently low temperatures and constant pressure $P$, the critical field data can be fitted to the thermodynamical relation[5,24]

$$H_c^2 = H_0^2 - 4\pi\gamma^* T^2 \quad (1)$$

where $H_0 = H_c(P, 0)$, $\gamma^* = \gamma/V$, $V$ is the molar volume at pressure $P$, and $\gamma$ is the temperature coefficient of the normal electronic specific heat at the same pressure. By using Eq. (1) for $P = 0$ and for $P \neq 0$ we get the relation

$$H_c^2(P, T) = H_c^2(P, 0) - \left(1 + \frac{\Delta\gamma^*}{\gamma^*}\right) \times [H_c^2(0, 0) - H_c^2(0, T)] , \quad (2)$$

where $\gamma^* = \gamma^*(P = 0)$ and $\Delta\gamma^* = \gamma^*(P) - \gamma^*(0)$. Equation (2) shows that the relation between the experimental points $H_c^2(P, T)$ and $H_c^2(0, T)$ is a straight line in the temperature range where Eq. (1) is valid. Recent measurements[25,26] of $H(0, T)$ of Pb show that this range extends to 2 K. Thus, $\partial \ln\gamma^*/\partial P$ was estimated directly from our experimental data using Eq. (2) and a least-squares fit, without relying on the considerably different values of $\gamma^*$ reported in the literature.[26]

The value of $H_c(P, 0)$ found using Eq. (2) and the straight-line fit is highly dependent on the published data of $H_c(0, 0)$.[21,25,26] The most reliable value is given by hysteresis-free high-accuracy experiment of Harris and Mapother (HM).[25] In contrast, the other measurements had either a high-hysteresis correction[21] or no correction at all.[26] To check the value of $H(P, 0)$ obtained by this method a second approach had been used. A polynomial fit was found for the 56 experimental points of HM in the range $0.35 < T < 4.2$ K. It turned out that the polynomial $H_c(0, T) = \sum_{n=0}^{N} a_n T^{2n}$, with $N = 4$, gave the best fit. Higher values of $N$ ($N \leq 8$) did not improve the result because of the experimental errors. This function was used as the temperature standard in the present experiment. By inverting the polynomial we have estimated the temperature $T$ for every experimental point. This temperature, together with experimental results $H_c(P, T)$ and $H_c(0, T)$ for the 3-kbar run are shown in Table I. A plot of $\partial H_c/\partial P$ vs $\bar{t}^2$ is shown in Fig. 2. Here $\partial H_c/\partial P = \Delta H_c(T)/P$, where $\Delta H_c(T) = H_c(P, T) - H_c(0, T)$, $P = 2960$ atm. and $\bar{t} = T/T_c(P = 0)$. It

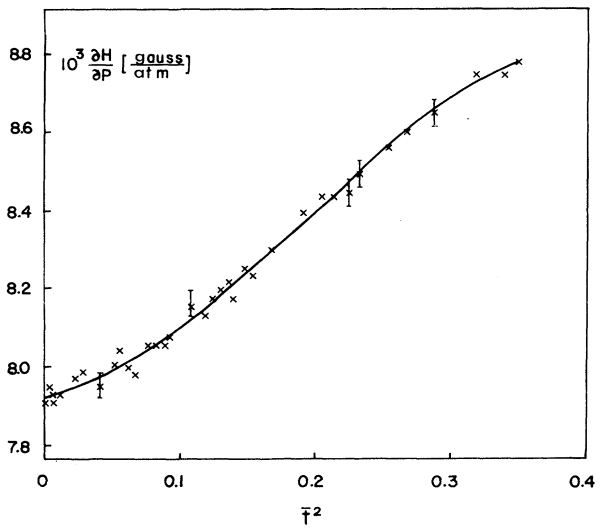


FIG. 2. Temperature dependence of $\partial H_c/\partial P$ for Pb vs $\bar{t}^2$ where $\bar{t} = T/T_c$ $(P = 0)$.

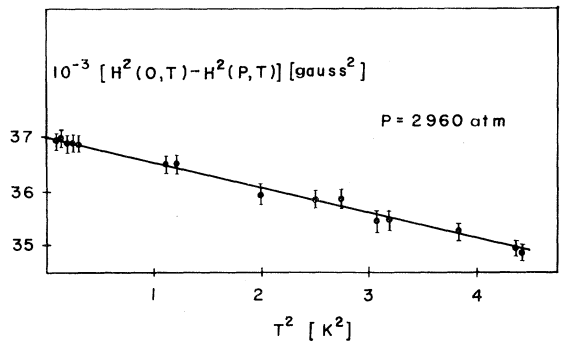


FIG. 3. The change in the critical magnetic field of lead $H_c^2(0,T) - H_c^2(P,T)$ due to a pressure of 2960 atm. vs $T^2$.

can be seen immediately from the graph that $\Delta H_c(T)$ changes less than the experimental error (0.1 G) for $t^2 < 0.01$ which corresponds to $T < 0.7$ K. The exact value of the temperature is not important as long as we are in the $He^3$ range. Thus $\Delta H_c(0)$ found by extrapolating Fig. 2 [or calculating $\Delta H(0)$ from the $He^3$ range points] is not dependent on the exact values of the polynomial constants. The numerical results obtained for $\Delta H(0)$ in this manner and by Eq. (2) do agree to within 0.1 G.

To cross check the results of both $\Delta H_c(0)$ and $\Delta\gamma^*$ we made use of Eq. (1) twice to get

$$H_c^2(0,T) - H_c^2(P,T) = H_c^2(0,0) - H_c^2(P,0) + 4\pi\Delta\gamma^* T^2 . \quad (3)$$

A plot of $H_c^2(0,T) - H_c^2(P,T)$ vs $T^2$ for low temperatures ($T < 2$ K) is shown in Fig. 3. In this case we made use of both $H_c(0,0)$ and $\gamma^*$ measured by HM. The results obtained for $\Delta H(0)$ and $\partial \ln\gamma^*/\partial P$ are equal to those previously found, within the experimental error.

The physical constants of superconducting lead used in this paper are shown in Table II. Those constants have been assumed as accurate values that do not contribute to the errors quoted in our final data. The results obtained in our experiments are summarized in Table III. The errors quoted for our results, especially those involving $H_0$, are due mainly to the uncertainty in the pressure measurement. For comparison, we have shown in Table III the experimentally measured parameters obtained by other methods

TABLE II. Physical constants of Pb (at $P = 0$).

| | |
|---|---|
| $T_c$ | 7.193 K[a] |
| $H_0$ | 801.48 G[b] |
| $\gamma$ | 2.972 mJ/mole K[c] |
| $V$ | 17.86 $cm^3$ [d] |
| $\partial T_c/\partial P$ | $-3.7\times10^{-5}$ K $atm^{-1}$ [e] |
| $K$ | $2.076\times10^{-6}$ $atm^{-1}$ [f] |

[a]J. D. Frank and D. L. Martin, Can. J. Phys. 39, 1520 (1961).
[b]Reference 25.
[c]Reference 25.
[d]Reference 21.
[e]Average of four experimental results Refs. 1, 8, 12, 27.
[f]D. L. Waldorf and G. A. Alers, J. Appl. Phys. 33, 3266 (1962).

TABLE III. Pressure and volume derivatives of $H_0$ and $\gamma^*$ of superconducting Pb.

| | $10^3 \frac{\partial H_0}{\partial P}\left(\frac{G}{atm}\right)$ | $\frac{\partial \ln H_0}{\partial \ln V}$ | $\frac{\partial \ln\gamma^*}{\partial \ln V}$ |
|---|---|---|---|
| This work | $-7.93 \pm 0.18$ | $4.77 \pm 0.12$ | $3.6 \pm 0.4$ |
| Ott[a] | $-8.0 \pm 0.15$ | $4.85 \pm 0.15$ | ••• |
| Collins and White[b] | ••• | ••• | $0.7 \pm 0.5$ |

[a]Reference 12. [b]Reference 4.

The general deviation function defined as

$$D(P,t) = H_c(P,T)/H_c(P,0) - (1 - t^2) , \quad (4)$$

where $t = T/T_c(P)$ was found experimentally at $P = 3$ kbar from our data using the temperature standard as above and at $P = 0$ from HM. $\partial T_c/\partial P$ [1,8,12,27] was taken from published data. However, as the reported values of $\partial T_c/\partial P$ vary as much as 15%, an average value was found, using the same compressibility $K$, for all the results (see Table II). The plot of $D(P,t)$ vs $t^2$ is shown in Fig. 4.

The shape of $D(P,t)$, while slightly dependent on $\partial T_c/\partial P$, is clearly changed by the 3 kbar pressure toward lower values. This observation shows that the similarity principle assumption used in previous papers is invalid and also shows directly that lead becomes a weaker-coupling superconductor as pressure is applied.

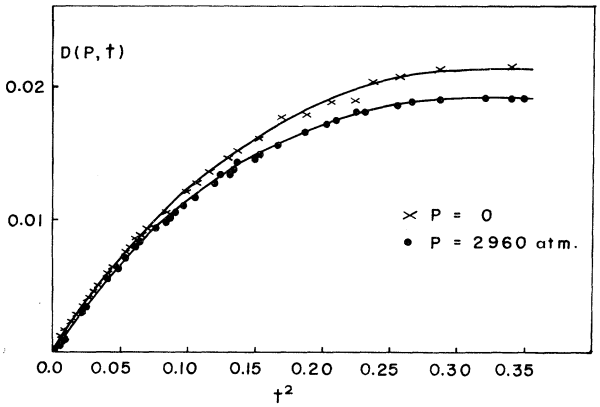


FIG. 4. Deviation of the critical field of Pb from parabolic temperature dependence $D(P,t)$ vs $t^2$ at two pressurss, where $t = T/T_c(P)$ is the reduced temperature.

TABLE IV. Comparison of $\partial \ln\lambda/\partial \ln V$ for superconducting Pb.

| Author | Method | $\partial \ln\lambda/\partial \ln V$ |
|---|---|---|
| This work | Critical field | 6.4 |
| Frank *et al.*[a] | Tunneling | 9.0 |
| Hansen *et al.*[b] | Tunneling | 3.7 |
| Carbotte and Vashista[c] | Theoretical | 4.8 |
| Boughton *et al.*[d] | Theoretical | 5.75 |

[a]Reference 9.
[b]An average for $Pb_{88}In_{12}$ and $Pb_{95}In_5$ from Ref. 11.
[c]References 3 and 7.
[d]Value obtained using Eq. (11).

### B. Strong-coupling parameters

Since Pb is a strong-coupling superconductor, it has been a subject of intense experimental and theoretical work, including pressure effects.[1-12] The main problem in the field is the unresolved discrepancy in the experimental results of $\partial \ln\gamma^*/\partial \ln V$ and $\partial \ln\lambda/\partial \ln V$. Here $\lambda$ is the electron phonon interaction parameter. The value of $\partial \ln\gamma^*/\partial \ln V$ obtained from thermal expansion[4] is in disagreement with $\partial \ln\gamma^*/\partial \ln V$ found by critical field measurements, including the present work (see Table III). These two methods are the only ones giving $\partial \ln\gamma^*/\partial \ln V$ directly. The value of $\partial \ln\lambda/\partial \ln V$[3,6-11] is measured directly by tunneling techniques. A comparison of the published data appears in Table IV, clearly showing the problem.

The electronic-specific-heat coefficient is given by

$$\gamma^* = \tfrac{2}{3}\pi^2 K^2 N(0)(1+\lambda) , \tag{5}$$

where $N(0)$ is the band-structure density of states at the Fermi surface per unit volume.[28] The volume dependence, given by

$$\frac{\partial \ln\gamma^*}{\partial \ln V} = \frac{\partial \ln N(0)}{\partial \ln V} + \frac{\lambda}{1+\lambda}\left(\frac{\partial \ln\lambda}{\partial \ln V}\right) , \tag{6}$$

was measured directly in this experiment. Assuming a free electron model,[3,7,28] i.e., $\partial \ln N(0)/\partial \ln V = -\frac{1}{3}$, the value of $\partial \ln\lambda/\partial \ln V$ appearing in Table IV was estimated, using $\lambda$ from Ref. 7.

The theoretical estimates of $\partial \ln\lambda/\partial \ln V$ were made using different approaches. In Ref. 7 the following scaling law was found:

$$\lambda(P) = (B/\beta^2)\lambda(0) , \tag{7}$$

where $B$ is a factor taking into account the average change under pressure of the matrix element of the electron-ion pseudopotential form factor and $\beta$ is a scaling factor of the phonon spectrum. Equation (7) was found assuming a single factor $\beta$ for the whole phonon spectrum and a constant value of $\alpha(\omega)$, where $\alpha$ is the electron-phonon coupling function. The theoretical calculation[6] shows that $B$ changes very little with pressure. Therefore a good approximation will be

$$\frac{\partial \ln\lambda}{\partial \ln V} = 2\gamma_G , \tag{8}$$

where $\gamma_G$ is the Grüneisen constant. This same result was obtained previously[29] from empirical relations. The numerical result, using $\gamma_G = 2.85$,[7] is $\partial \ln\lambda/\partial \ln V = 5.7$. The other theoretical approach[28] follows an approximation to McMillan's formula for $T_c$[30]:

$$T_c = \Theta_D e^{-1/g} \tag{9}$$

and

$$g = (\lambda - \mu^*)/(1+\lambda) . \tag{10}$$

Here $\Theta_D$ is the Debye temperature and $\mu^*$ is the Coulomb-interaction factor. Combining Eqs. (9–10) and differentiating with respect to volume we find

$$\frac{\partial \ln\lambda}{\partial \ln V} = \frac{(\lambda-\mu^*)^2}{\lambda(1+\mu^*)}\left(\frac{\partial \ln T_c}{\partial \ln V} + \gamma_G\right) . \tag{11}$$

To evaluate this we take $\lambda$ and $\mu^*$ from Ref. 7 and $\partial \ln T_c/\partial \ln V$ from Table II ($\lambda = 1.53$, $\mu^* = 0.15$) to obtain $\partial \ln\lambda/\partial \ln V = 5.75$, a value nearly equal to our former estimate and to our experimental result (see Table IV).

It should be point out that the comparison between the theory[7,29] which estimates $\partial \ln\lambda/\partial \ln V$ and an experiment which gives $\partial \ln\gamma^*/\partial \ln V$ assumes the free-electron model. Although this comparison is widely used[3,7,11,12] it should be treated with extreme caution. Another common computation was to find $\partial \ln\gamma^*/\partial \ln V$ from $\partial \ln T_c/\partial \ln V$ and $\partial \ln H_0/\partial \ln V$. At very low temperatures[3]

$$H_c(P,T) = H_c(P,0)[1-(1-\alpha)t^2] , \tag{12}$$

where $1-\alpha = 2\pi\gamma^* T_c^2/H_c^2(P,0)$.

Carbotte[3] has found theoretically that $\partial \ln(1-\alpha)/\partial \ln V = -2$. Using this value, any experiment[1,12] giving the pressure derivatives of $T_c$ and $H_0$ could be used to find $\partial \ln\gamma^*/\partial \ln V$. However, there is a large theoretical correction to the experimental data. Furthermore, a value of $\partial \ln(1-\alpha)/\partial \ln V = -1$ was estimated in this work using our experimental values from Table III and $\partial T_c/\partial P$ from Table II.

## IV. CONCLUSION

To summarize the present situation, the experimental parameters $\gamma^*$, $\lambda$, $H_0$, and $T_c$ of Pb are known to a reasonably good accuracy. As for the experimental volume derivatives: the quantities $\partial \ln H_0/\partial \ln V$ found by two different methods are the same, the quantities $\partial \ln T_c/\partial \ln V$ have a 15% spread,

and the results for $\partial \ln\gamma^*/\partial \ln V$ and $\partial \ln\lambda/\partial \ln V$ obtained by different authors are in disagreement, even for experiments done by the same method. In this work a better estimate for $\partial \ln\gamma^*/\partial \ln V$ was made and the value for $\partial \ln H_0/\partial \ln V$ was firmly established.[12] To confirm our result for $\partial \ln\gamma^*/\partial \ln V$, a thermal expansion measurement of the same parameter is highly desirable; the only existing measurement is in complete disagreement with both theory and this experiment. To complete our knowledge on superconducting lead, more experiments are needed to find an accurate value of $\partial \ln\lambda/\partial \ln V$ and to estimate $\partial \ln N(0)/\partial \ln V$. In this way, a complete meaningful comparison of all the superconducting strong-coupling parameters of lead will be possible.

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