p. 37.

 $37E.$ A. Lynton, in Ref. 36, p. 40.

³⁸D. E. McCumber, J. Appl. Phys. 40, 3113 (1968).

³⁹J. Clarke and T. A. Fulton, J. Appl. Phys. 40, 4470

(1969).

⁴⁰H. A. Atwater, Introduction to Microwave Theory

(McGraw-Hill, New York, 1962), p. 222.

 $⁴¹P$. L. Richards, *Physics of III-V Compounds* (Academic,</sup> New York, 19XX), Vol. 6.

 42 B. T. Ulrich and E. O. Kluth, in Proceedings of the Applied Superconductivity Conference, Annapolis, Md., 1972 (unpublished).

³T. D. Clark, Phys. Lett. A 27, 585 (1968).

⁴⁴D. J. Repici, L. Leopold and W. D. Gregory, in Ref. 42.

⁴⁵E. Riedel, Z. Naturforsch. A 19, 1634 (1964).

N. R. Werthamer, Phys. Rev. 147, 255 (1966); see S. A.

Buckner, T. F. Finnegan, and D. N. Langenberg [Phys. Rev. Lett.

28, 150 (1972)] for experimental data.
⁴⁷J. Barden, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).

48B. W. Roberts, Superconducting Materials and Some of Their Properties, G. E. Res. Report No. 61-RL-2744M, June, 1961 (unpublished).

⁹T. D. Clark, in Proceedings of the Conference on Science of Superconductivity, Stanford, 1969 (ubpublished); Physica

(Utr.) 55, 432 (1971).

 C. C. Grimes, P. L. Richards, and S. Shapiro, Phys. Rev. Lett. 17, 431 (1968).

⁵¹C. C. Grimes, P. L. Richards, and S. Shapiro, J. Appl. Phys. 39, 3905 (1968).

²P. L. Richards and S. A. Sterling, Appl. Phys. Lett. 14, 394 (1969).

B. D. Josephson, Phys. Lett. 1, 251 (1962).

⁵⁴T. D. Clark, in Ref. 24, p. 449.

- ⁵⁵W. H. Parker, D. N. Langenberg, A. Denenstein, and B. N. Taylor, Phys. Rev. 177, 639 (1969).
	- '6M. J. Stephen, Phys. Rev. 186, 393 (1969).

57P. E. Gregers-Hansen, M. T. Levinsen, and G. Fog

- Pedersen, J. Low Temp. Phys. 7, 99 (1972).
	- $58P$. Russer, Acta Phys. Austriaca 32, 373 (1970).

⁵⁹H. Fack and V. Kose, J. Appl. Phys. **42,** 320 (1971).

⁶⁰J. Crow and M. Strongin (private communication).

⁶¹R. De Bruyn Ouboter and A. Th. A. M. DeWaele, Prog.

- Low Temp. Phys. 6, 273 (1970). $62P$. G. de Gennes, Superconductivity of Metals and Alloys
- (Benjamin, New York, 1966), p. 10.
	- ³J. Kurkijärvi and W. W. Webb, in Ref. 42. 6'T. Finnegan and S. Wahlsten, in Ref. 4.
	-

6'S. Shapiro, J. Appl. Phys. 38, 1879 (1967).

PHYSICAL REVIEW B VOLUME 8, NUMBER 1

1 JULY 1973

Magnetic Measurements on Strong-Coupling $Pb_{1,2x}Bi_xTl_x$ and Pb-In Superconducting Alloys^{*}

J. H. Fearday[†] and R. W. Rollins Department of Physics, Ohio University, Athens, Ohio 45701 (Received 25 September 1972)

Measurements are presented of the generalized Ginzburg-Landau parameter $\kappa_2(T)$ and the bulk upper-critical field $H_{c2}(T)$ as a function of temperature T near the critical temperature in Pb, $\partial_{r}B_{i\sigma}T_{i\sigma}$, where $x = 0.015, 0.05, 0.10,$ and 0.15, and of $H_{c2}(T)$ in Pb₁, In_y, where $y = 0.018, 0.53, 0.087,$ and 0.167. These results, and previous measurements of κ_2 in Pb-In by Farrell et al., which are in disagreement with weak-coupling theories are found to be in ageement with calculations by Eilenberger and Ambegaolar and by Usadel which include both strong-coupling and electron-mean-free-path effects.

I. INTRODUCTION

We present in what follows measurements of the generalized Ginzburg-Landau¹⁻⁴ parameter κ_2 and the upper-critical field H_{c2} as a function of temperature for some Pb alloys for which the elec-'tron-phonon coupling is too strong to be treated by the weak-coupling extensions^{2,3} of the Bardeen Cooper-Schrieffer (BCS)⁵ theories. The measurements are shown to be in general agreement with recent calculations, which include both strongcoupling and electron-mean-free-path effects, by Elienberger and Ambegaokar 6 and by Usadel.

Elienberger and Ambegaokar have shown that when calculating $(dH_{c2}/dT)_{T_c}$, the effects of strongcoupling and of electron mean free path may be factored. The strong-coupling factor does not depend explicitly on the mean free path and is expressed in terms of the thermodynamic critical

field $H_c(T-T_c)$ and the energy-gap parameter $\Delta(T - T_c)$, which may be obtained from experiment. Usadel. has shown a similar strongcoupling factor exists for $\kappa_2(T_c)$ in the dirty limit.

The Pb-Bi- Tl system was chosen for several reasons: (a) Disordered substitutional alloys having the Pb crystal structure exist over a wide range of compositions, simplifying sample preparation; (b) all the constituents have nearly the same mass and, therefore, changes in the phonon spectrum and electron-phonon coupling due to ionic-mass changes should be minimized; (c) the $Pb_{1-2x}Bi_xTl_x$ system has a constant electron-to-atom ratio of 4, thus creating a system of "artifical lead" with essentially constant phonon spectrum and constant electrongas density, giving rise to a constant electronphonon coupling for the entire system; (d) tunneling measurements have been reported by Dynes and Rowell⁸ for $Pb_{1-2x}Bi_xTl_x$ alloys.

162

The Pb-In system was studied because previous measurements by Farrell et al.⁹ have shown discrepancies between measured and calculated values of $\kappa(T_c)$, and because the necessary tunneling data have been reported by Adler et $al.$ ¹⁰

II. EXPERIMENTAL METHODS

A. Sample Preparation

 $Pb_{1-2x}Bi_xTl_x$ alloys with $x = 0.015, 0.05, 0.10,$ and 0.15, and Pb_{1-y} In_y alloys with $y = 0.018$, 0.053, 0. 087, and 0. 167 were prepared for investigation. All samples were polycrystalline cylinders $\frac{1}{8}$ in. in diameter and 2 in. long. The 99.999% pure lead, bismuth, and thallium starting metals were weighed, melted, thoroughly mixed, cast into $\frac{1}{2}$ -in. diam cylinders and then quenched. The entire procedure was done in an evacuated Pyrex container using care to avoid contamination. The Pb-In system employed 99. 999% pure lead and indium as starting materials. The metals were melted, mixed, and quenched in a $\frac{1}{2}$ -in. -diam Pyrex tube. The $\frac{1}{2}$ -in. -diam ingot was then extruded into $\frac{1}{8}$ -in. diam samples. As a check to see whether results obtained fromextruded samples are the same as results obtained from cast samples, a $Pb_{.90}Bi_{.05}Ti_{.05}$ sample was extruded in the same manner as the Pb-In samples and the results compared with those of the cast $Pb_{.90}Bi_{.05}T1_{.05}$ sample. The two samples showed no significant differences either in their values of T_c or $\kappa_2(t)$. After each sample was made and cut into a length of 2 in., it was sealed in an evacuated Pyrex tube and annealed for periods of time ranging from one to three weeks at a temperature within ⁵ to 10 'C below the solidus line.

B. Temperature Measurement and Control

The temperature measurement and control for both ac and dc magnetization measurements was accomplished in the same manner. The temperature was measured using calibrated CryoCal¹¹ germanium resistance thermometers on each of the probes. The calibrations of the two thermometers were checked against each other and by measuring the critical temperature of 99. 999% pure Pb to be 7.205 ± 0.010 K, which compares with 7. 193 \pm 0.005 K reported by Frank and Martin.¹² Below 4. 2 K the calibrations were checked against the $He⁴$ -vapor-pressure scale.

Temperature control above \sim 4 K was accomplished by using an electronic regulator with a carbon resistance thermometer as a sensor and a wirewound heatex. The sample, pick-up coils, and thermometers were all kept in thermal contact by means of a copper sample holder. This sample holder was suspended from a small stainless-steel tube and was otherwise isolated from the external helium bath by a vacuum jacket. The temperature of the external helium bath was always kept below the desired sample temperature. Heat flowing out of the sample and sample holder through the stainless-steel suspension tube was then controlled by the wire-wound heater which was at the base of this tube. Below \sim 4 K the sample and holder were immersed in liquid $He⁴$ and the temperature controlled by pumping on the liquid helium. Temperatures in the range of 1.5 to 8 K could be controlled to 0.001 K in this manner.

C. Magnetic and Resistivity Measurements

The ac permeability was measured by applying a 210-Hz small-amplitude ac field with a primary coil and measuring the voltage induced in the secondary coil containing the sample. The primary and secondaxy coils were coaxial and measured the middle $\frac{1}{2}$ in. of the sample. A lock-in detector was used to detect the induced voltage giving the in-phase and out-of-phase permeabilities $(\mu'$ and μ''), as previously described.¹³

The dc magnetization measurements were carried out using a similar probe with the secondary coil replaced by two 30000-turn coils wound with No. 48 copper wire, one containing the sample. The difference voltage induced in these coils by a slowly changing external dc field was electronically integrated using a system similar to that described by Fietz¹⁴ and giving the magnetization directly on the Y axis of an $X-Y$ plotter. The X axis was driven by the integrated induced voltage of the empty coil and thus was proportional to the applied field.

A superconducting solenoid provided the applied dc magnetic fields for measurements above 1 kOe, the current-field characteristic being determined by calibration with NMR. This characteristic was found to be reproducible within 0. 5% at fields above 1 kOe. Below about 1 kOe, the field was supplied by a sixth-order liquid-nitrogen-cooled solenoid which was free from ac ripple, noise, and hysteresis for work near T_c . The current-field characteristic of this solenoid was also determined by NMR and allowed the field to be determined to within 1% .

The dc resistivity of all samples was measured just above the critical temperature using a standard four-lead method.

D. Determination of T_c

 T_c was obtained by measuring the in-phase component of the permeability μ' , using an ac-field amplitude of 0. 004 Oe. The permeability was found to be independent of ac-field amplitude for small amplitudes. The in-phase permeability μ' goes from 0 to 1 as the temperature is increased through the transition temperature T_c . The center of the transition is taken to be T_c . The transitions had a half-width of about 0. 005 K, which represents

Sample	$T_c(K)$ (± 0.010)	$\rho_n(\mu\Omega)$ cm) $(\pm 3\%)$	$f(\rho)$	$2\Delta^{\text{expt}}(0)$ $k_B T_c$	$\frac{dH_c}{dT}$ (Oe/K) T_c	Measured $\frac{dH_{c2}}{dT}$ (Oe/K) T_c $(+1%)$	Calculated $\frac{dH_{c2}}{dT}$ (Oe/K) T_{c} _(+ 15%)
P _b				4.36 ²	238		
$Pb_{0.982}$ In _{0.018}	7.143	1.43	1.391	4.42^a	239	237	210
$Pb_{0.947}$ In _{0.053}	7.086	4.24	1.064	4.42 ^a	239	473	480
Pb_0 913 $In_{0.087}$	7.040	5.87	1.015	4.42 ^a	239	696	640
$Pb_{0.833}$ In _{0.167}	6.929	10.8	0.951	4.42^a	239	1124	1090
$Pb_{0.97}Bi_{0.015}Tl_{0.015}$	7.204	2.29	1.21	4.38 ^b		310	300
$Pb_{0.90}Bi_{0.05}Tl_{0.05}$	7.278	7.62	0.982	4.37 ^b		865	810
$Pb_{0.80}Bi_{0.10}Tl_{0.10}$	7.344	15.62	0.924	4.42 ^b		1637	1530
$Pb_{0.70}Bi_{0.15}Tl_{0.15}$	7.376	23.04	0.903	4.46 ^b		2483	2160

TABLE I. Experimental and theoretical values of $(dH_{c2}/dT)_{T_c}$ calculated from other parameters listed as described in the text. The $\pm 15\%$ error assigned to the calculated values of $(dH_{c2}/dT)_{T_a}$ includes $\pm 8\%$ possible systematic error in $2\Delta^{\text{expt}}/k_BT_c$ suggested by the different values obtained for pure Pb in Refs. 8 and 10.

^aFrom Ref. 10.

^bFrom Ref. 8 multiplied by a factor such that $2\Delta/k_BT_c = 4.36$ for pure Pb.

the temperature interval in which μ' increase from $\frac{1}{2}$ to $\frac{3}{4}$ of its normal value. A check of T_c was also made using the dc magnetization measurements. This is determined by approaching T_c from below and noting the temperature at which the magnetization goes to zero as the external field is swept above and below zero. This determination of T_c was accurate to within 0.005 K and agreed with the ac determination.

The T_c results are shown in Table I.

III. RESULTS AND DISCUSSIONS

A. Determination of $\kappa_2(T)$ for Pb-Bi-Tl System

The experimental quantity $\left[dM(T)/dH\right]_{H_{c2}}$ is related to the generalized Ginzburg-Landau parameter $\kappa_2(T)$ by the equation³

$$
\left(\frac{dM(T)}{dH}\right)_{H_{c2}} = -\left\{4\pi\beta \left[2\kappa_2^2(T)-1\right]\right\}^{-1}.
$$
 (1)

For this work a triangular fluxoid lattice, $\beta = 1.16$, is assumed.¹⁵ A typical dc magnetization curve is shown in Fig. 1. Altogether about 20 different isothermal magnetization curves were obtained at temperatures ranging from 0.15 T_c to 0.98 T_c for each of the Pb-Bi- Tl samples.

As can be seen from Fig. 1, it is not permissible to ignore the hysteresis. In order to obtain $\left(dM/dH \right)_{H_{c2}}$, the slope of the magnetization curve at H_{c2} was taken for both increasing and decreasing fields. The average of these slopes was then used to calculate the values of $\kappa_2(t)$ shown in Fig. 2. The error bars show the extremes due to calculating $\kappa_2(t)$ from the increasing and decreasing slopes. For the $Pb_{0.70}Bi_{0.15}Tl_{0.15}$ sample, $(dM/dH)_{Hc2}$ became small, so that the very small slope of $\left(dM/dH \right)_{Hc2}$ in a decreasing field gave rise to a

very large $\kappa_2(t)[\kappa_2(t) - \infty$ as $(dM/dH)_{H_{c2}} \to 0]$. Errors in $\kappa_2(t)$ therefore became large for the higher samples. Errors in $\kappa_2(t)$ due to a nonzero demagnetization coefficient, instrumentation errors, and errors in determining the slopes, were less'than errors in determining the slopes, were less that
~2%, and were therefore small compared to uncertainties introduced by the irreversibility just discussed.

The measured value of $\kappa_2(T_c)$ was determined by extrapolating $\kappa_2(T)$ to $T = T_c$. The results are shown in Table II for all but the $x = 0$. 15 sample, where errors were prohibitively large.

B. Determination of $(dH_{c2}/dT)_T$.

Values of $H_{c2}(T)$ for $T \le 0$. $9T_c$ were obtained from the dc magnetization curves as indicated in Fig. 1. These values could be determined to an accuracy of 1-2%. For $T > 0.9T_c$, the dc fields become so small that the dc drift in the magnetometer becomes too large for an accurate determination. Therefore the values of H_{c2} near T_c were taken from ac permeability data. The determination of H_{c2} from this type of data has been extensively discussed elsewhere.¹³ H_{c2} coincides with a break in the slope of both the in-phase and out-of-phas permeabilities μ' and μ'' as a function of applie h the in-
and μ'' dc field at constant temperatures. This break in the slope occurs provided the ac-field amplitude is large enough to penetrate the surface sheath, The ac-field amplitude used was on the order of $10^{-3}H_{c2}$. The values of H_{c2} determined from this method are precise to better than 1%. Values of H_{c2} obtained from the dc and ac methods give the same values in their region of overlap.

The measured values of $(dH_{c2}/dT)_{T_c}$ listed in Table I were obtained from the slope of a $H_{c2}(t)$ vs t plot as shown in Fig. 3.

FIG. 1. Typical magnetization curve.

C. Determination of ρ and κ ^{BCS} for Pb-Bo-Tl Alloys

The Górkov impurity parameter ρ can be calculated from²

$$
\rho = 8.85 \times 10^{-3} \gamma^{1/2} \rho_n / \kappa_0, \qquad (2)
$$

where the residual resistivity ρ_n has been measured and is shown in Table II. γ is the electronic specific-heat coefficient in ergs cm³ K⁻², and κ_0 is the Qinsburg-Landau parameter in the clean limit. The renormalized Ginsburg-Landau k can then be expressed in terms of ρ through the Górkov impurity function $\chi(\rho)$ as

$$
\kappa^{\rm BCS} = \kappa_0 / \chi(\rho) \tag{3}
$$

where κ^{BCS} is a renormalized BCS parameter and where

$$
\chi(\rho) = 0.951 \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2 (2n+1+\rho)} \quad . \tag{4}
$$

The expression for κ_0 given by Berlincourt and Hake 16 is

$$
\kappa_0 = 1.61 \times 10^{24} T_c \gamma^{3/2} [N_e^{4/3} (S/S_0)]^{-1} . \tag{5}
$$

In this expression N_e is the effective conductionelectron density in electrons/ cm^3 , S is the area of the Fermi surface, and S_0 is the area of the Fermi surface in the free-electron model. If this equation is normalized by the pure-lead values, and we assume that S/S_0 is the same for all the alloys as for pure lead, then

$$
\kappa_0 = (\kappa_0)_{\rm Pb} [\gamma / \gamma_{\rm Pb}]^{3/2} [N_e / (N_e)_{\rm Pb}]^{-4/3} T_e / (T_c)_{\rm Pb} . (6)
$$

For the $Pb_{1-2x}Bi_xTl_x$ system, the number of electrons per atom is constant, and the atomic density is a constant to 0.2%, so that $N_e \simeq (N_e)_{\text{Pb}}$. Also, since the electron density and the electron-phonon coupling are nearly constant for the $Pb_{1-2x}Bi_xTI_x$ system, γ should be nearly the same for the alloys as for pure lead. Making these assumptions and using $(T_c)_{\text{Pb}} = 7.205 \text{ K}$, $\gamma_{\text{Pb}}^9 = 1.69 \times 10^3 \text{ ergs cm}$
 K^{-2} , and $(\kappa_0)_{\text{Pb}}^1 = 0.24 \pm 0.007 \text{ gives}$

$$
\kappa_0 = 0.0333T_c, \qquad (7)
$$

$$
\rho = 10.92 \rho_n / T_c \tag{8}
$$

Using measured values of ρ_n and T_c in Eqs. (4), (7), and (8), values of κ^{BCS} are calculated from Eq. (3) and shown in Table II along with the experimental values of $\kappa_2(T_c)$. Values of κ_0 and $\chi(\rho)$ are also tabulated. As can be seen, the values of κ^{BCS} are about 15% higher than the measured values of $\kappa_2(T_c)$.

D. Theroetical Determination of κ_2 Including Strong Coupling and Comparison with Experiment

Equation (3) is based on the weak-coupling theories. Usadel has recently calculated the strongcoupling correction to $\kappa_2^{\text{BCS}}(T_c)$ in the dirty limit $\rho \rightarrow \infty$. He finds

$$
\kappa_2^{\rm SC} \equiv \kappa_2^{\rm SC}(T_{\rm c}) = \Lambda_{\kappa} \kappa_2^{\rm BCS}(T_c) \quad , \tag{9}
$$

where $\kappa_2^{\rm SC}(T_c)$ and $\kappa_2^{\rm SC}$ are values which have been corrected for strong coupling. The strong-coupling factor Λ_{κ} is given by

$$
\Lambda_{\kappa} = \left[\frac{dH_c^{\text{expt}}/dT}{dH_c^{\text{BCS}}/dT} \left(\frac{\Delta^{\text{BCS}}}{\Delta^{\text{expt}}} \right)^2 \right]_{T_c} . \tag{10}
$$

The terms dH_c^{expt}/dT and Δ^{expt} are the experimentally observed values of these parameters. Equation (3) therefore becomes

$$
\kappa^{\rm SC} = \Lambda_{\kappa} \, \kappa_0 / \chi(\rho) \; . \tag{11}
$$

FIG. 2. $\kappa_2(t)$ vs $t \equiv T/T_c$. The error bars show the extremes due to calculating $\kappa_2(t)$ from the increasing and decreasing slope $\left(\frac{dM}{dH}\right)_{H_{c2}}$.

TABL" II. *K* parameters for the Pb-Bi-Tl system. The errors shown for each of the parameters are total errors resulting from errors in experimental parameters. The $\pm 9\%$ error for $\kappa_2^{\rm SC}$ is due to the $\pm 4\%$ error in $\kappa^{\rm BCS}$ plus an estimated possible 5% systematic error in the pure-Pb parameters used in calculating the strong-coupling factor.

Sample	κo $(\pm 3\%)$	ο (± 6%)	$\chi(\rho)$	κ _{BCS} $(\pm 4\%)$	Measured $\kappa_2(T)$	Calculated $\kappa_2^{\rm SC}(T_c)$ $(\pm 9\%)$
$Pb_{0.97}Bi_{0.015}Tl_{0.015}$	0.240	3.47	0.2380	1.01	0.91 ± 0.08	0.908
$Pb_{0.90}Bi_{0.05}Ti_{0.05}$	0.242	11.43	0.0891	2.69	2.41 ± 0.15	2.42
$Pb_{0.80}Bi_{0.10}Ti_{0.10}$	0.245	23.23	0.0466	5.15	4.73 ± 0.3	4.63

As a first approximation, the strong-coupling factor Λ_{κ} will be assumed to be the same for all the alloys as for pure lead. This assumption will be discussed later and shown to hold to within a few percent. The pure-lead values which enter into the correction factor are $(-dH_c^{\text{expt}}/dT)_{T_c} = 238$ Oe/K , ¹⁸ ($\Delta^{\text{expt}}/\Delta^{\text{BCS}}|_{T_a}^2 = 1.53$, and $\left(-dH_c^{\text{BCS}}/dT\right)$ $= 173$ Oe/K.⁶ Equation (11) then becomes

$$
\kappa^{\rm SC} = (0.899 \pm 0.045) \, \kappa_0 / \chi(\rho). \tag{12}
$$

Using values of κ_0 and $\chi(\rho)$ from Table I, values of κ^{SC} are calculated and shown. These values of κ^{SC} are well within the experimental limits of $\kappa_2(T_c)$.

Recent experimental results on Pb-In alloys also show a discrepancy between the measured and BCS values of κ . These values are shown in Table III. The strong-coupling correction factor for these alloys is also constant to within a few percent and equal to the pure-lead correction. Using Eq. (12), values of κ^{SC} are calculated for the Pb-In system and shown in Table III. These corrected values bring the theoretical and experimental values of κ into better agreement for the Pb-In samples with less than 30-at. $%$ In. For the higher percentage alloys, much better agreement is obtained, but a discrepancy still remains. Some evidence has been shown⁹ to indicate a modification in the electronic structure in the Pb-In system at 30-at. % In which may be responsible for the remaining disagreement.

E. Calculation of $(dH_{c2}^{SC}/dT)_{T_c}$ and Comparison with Experimer

Eilenberger and Ambegaokar 6 have calculated the strong-coupling correction term to the BCS value of $(dH_{c2}/dT)_{T_c}$ in terms of measurable quantities. According to their calculations,

$$
\left[\frac{dH_{c2}^{\rm SC}(T)}{dT}\right]_{T_c} = \Lambda_{H_{c2}} \left[\frac{dH_c^{\rm BCS}(T)}{dT}\right]_{T_c} \quad , \tag{13}
$$

where $\Lambda_{H_{c2}}$ is the strong-coupling factor given by

$$
\Lambda_{H_{c2}} = \left[\frac{dH_c^{\text{expt}}/dT}{dH_c^{\text{DCS}}/dT} \frac{\Delta^{\text{BCS}}}{\Delta^{\text{expt}}} \right]_{T_c}^2. \tag{14}
$$

The renormalized BCS quantities in Eq. (13) can be written as

$$
\left[\frac{dH_{c2}^{\text{BCS}}}{dT}\right]_{T_c}^2 = 2.36 \,\chi^{-1}(\rho) \,\frac{T_c}{k_F^2} \left[\gamma S_0 / \gamma_0 S\right]^2 \,\text{Oe/K} \,\text{\AA} \tag{15}
$$

and

$$
\left[\frac{dH_c^{\text{BCS}}}{dT}\right]_{T_c}^2 = 0.93 \times 10^4 k_F (\gamma/\gamma_0) \text{ Oe}^2 \text{ Å/K}, \quad (16)
$$

where k_F is the Fermi wave number in \AA^{-1} and γ_0 is the free-electron specific-heat coefficient.

A new function $f(\rho)$ is now introduced which is defined by

$$
\chi^{-1}(\rho) \equiv f(\rho) \rho \tag{17}
$$

where $f(\rho)$ is evaluated from Eq. (4). Since $\chi^{-1}(\rho)$ is very nearly proportional to ρ for large ρ , $f(\rho)$ is a weakly dependent function of ρ for all of our sam-

FIG. 3. $H_{c2}(t)$ vs $t \equiv T/T_c$ for the Pb_{0, 97}Bi_{0, 015}Tl_{0, 015} sample as determined from both dc magnetization data and ac permeability data.

TABLE III. κ parameters for the Pb-In system. κ_0 and ρ values are calculated using Eqs. (2) and (6) from the text with $(\kappa_0)_{\text{Ph}} = 0.24 \pm 0.007$ and the values of γ and T_c for Pb-In alloys reported in Ref. 9. The errors in the parameters are the same as those in Table II.

Sample	κo (± 3%)	ρ $(* 6\%)$	κ _{BCS} $(\pm 4\%)$	measured $\kappa_2(T_\alpha)^a$ $(\pm 10\%)$	calculated $\kappa_2^{\rm SC}(T_c)$ $(\pm 9\%)$
$Pb_{0.95}$ In _{0.05}	0.243	5.11	1.38	1.25	1.24
$Pb_{0.90}$ In _{0.10}	0.247	9.33	2.31	2.17	2.08
$\rm Pb_{0.80} In_{0.20}$	0.253	15.96	3.83	3.50	3.44
$\rm Pb_{0.70} In_{0.30}$	0.261	22.07	5.34	4.22	4.80
$Pb_{0.60}$ In _{0.40}	0.267	24.34	5.99	4.52	5.39
$Pb_{0.30}$ In _{0.50}	0.275	24.17	6.12	4.62	5.50
$Pb_{0.60}$ In _{0.40}	0.282	21.04	5.52	4.15	4.96

aFrom Ref. 9.

ples. Therefore, any errors in ρ are only weakly present in the values of $f(\rho)$.

It is also necessary to relate $\Delta^{BCS}(T_c)/\Delta^{expt}(T_c)$ to measurable quantities. Since $\Delta(T)/\Delta(0)$ vs T/T_c follows the BCS curve very closely for pure Pb, 19 we assume

$$
\frac{\Delta^{\text{BCS}}(T_c)}{\Delta^{\text{expt}}(T_c)} = \frac{\Delta^{\text{BCS}}(0)}{\Delta^{\text{expt}}(0)} \quad . \tag{18}
$$

Since $\Delta(0)$ varies by less than 5% for both of our alloy systems, Eq. (18) is assumed to hold for both systems.

Substituting Eqs. (2) , (6) , (14) – (18) into Eq. (13) . it becomes

$$
\left[\frac{dH_{oz}^{SC}(T)}{dT}\right]_{T_c} = 0.0365 \left[\frac{2\Delta^{expt}(0)}{k_B T_c}\right]^{-2}
$$

$$
\times \rho_n f(\rho) \left[\frac{dH_c^{SC}(T)}{dT}\right]_{T_c}^2. \tag{19}
$$

So, by introducing $f(\rho)$, the parameters S/S_0 γ/γ_0 , and k_F all cancel out of Eq. (13). Equation (19)is then the strong-coupling prediction in terms of measurable quantities, with the exception of $f(\rho)$, which is a slowly varying function.

For a comparison with the preceding theoretical predictions, the values of T_c , $\Delta^{\text{expt}}(0)$, ρ_n , and $(dH_c^{\text{expt}}/dT)_{T_c}$ are needed. In this experiment ρ_n and T_c were measured for both sets of alloys. For the Pb-In alloys, $\Delta^{\text{expt}}(0)$ was determined from specific-heat measurements.²⁰ The measured values are shown in Table III and are used in Eq. (19) to determine the calculated strong-coupling values of $(dH_{c2}^{SC}/dT)_{T_c}$. These calculated values of $(dH_{c2}^{8}C/dT)_{T_c}$, along with the values of $(dH_{c2}/dT)_{T_c}$ measured in this experiment, are shown in Table I. As can be seen, $2\Delta^{\text{expt}}(0)/k_B T_c$ is a constant for the range of alloys considered, indicating that the coupling is essentially constant. Likewise, $(dH_c^{\text{expt}}/dT)_{T_c}$ is constant, so that according to Eq. (19), $(dH_{c2}^{SC}/dT)_{T_c} \propto f(\rho)\rho_n$. The calculated and

measured values of $(dH_{c2}/dT)_{T_c}$ show a maximum difference of 10%, and are, in general, within $5%$ over the range of alloys. For the Pb-Bi-Tl system $\Delta^{\text{expt}}(0)$ was also measured by electron tunneling. 8 These tunneling data, however, give a pure-lead value of $2\Delta^{SC}(0)/k_B T_c = 4.52$. This value is in disagreement with the value of 4. 36 found in the Pb-In measurements. Therefore, the values of $2\Delta^{SC}(0)/k_BT_c$ for the Pb-Bi-Tl system have been renormalized so that the pure-lead values agree for the two systems. These renormalized values are shown in Table I. $(dH_c^{\mathcal{SC}}/dT)_{T_c}$ is in the process of being measured, 21 but to a first approximation it is assumed to be constant and equal to the pureit is assumed to be constant and equal to the pure
lead value of 238 Oe/ \degree K.¹⁸ This assumption gives a value of $(dH_{c2}^{SC}/dT)_{T_c}$ in agreement with the measured value to within 3% for the Pb_{0.97}Bi_{0.015}Tl_{0.015} sample. The higher-impurity samples show progressively worse agreement. This remaining disagreement may be due to the assumption that $(dH_c^{\text{expt}}/dT)_{T_c}$ is a constant. From Eq. (16), which neglects strong coupling, it is expected that (dH_c^{expt}) $\left(dT\right)_{T_{_{\cal C}}}^2$ is proportional to γ times a strong-coupling term. Since the strength of the coupling, as indicated by $2\Delta(0)k_BT_c$, is increasing for increasing impurity concentration, both γ and the strong-coupling term should cause an increase in $(dH_c^{\text{expt}}/dT)_T$ of 5% between the pure-lead value and that of the highest impurity alloy, which would be sufficient to bring the measured and calculated values of $(dH_{c2}/dT)_{T_c}$ into agreement for the Pb-Bi-Tl system. This 5% increase can be shown to cause little or no increase in the value of $\kappa^{\rm sc}$ in Table II.

IV. SUMMARY AND CONCLUSIONS

The following conclusions may be drawn from this work:

(i) The strong-coupling correction factor to the Ginzburg-Landau parameter κ , which has been calculated by Usadel⁷ in the dirty limit may be applied with success to the general mean-freepath case. The correction factor brings about agreement between the measured and calculated values of κ for the Pb-Bi-Tl system, with Gorkov impurity parameter ρ ranging from 3.5 to 23, and for the Pb-In system for less than 30 -at. $%$ In in Pb. For alloys with a higher In concentration, a possible explanation for the remaining disagreement may be due to a band-structure change⁹ above the 30-at. % In composition.

(ii) The strong-coupling correction to the uppercritical field at T_c , as calculated by Eilenberger and Ambegaokar⁶ for the general mean-free-path case, is found to bring the measured and calculated values of $(dH_{c2}/dT)_{T_c}$ into agreement for the Pb alloys studied. Without the correction for strong coupling, the disagreement is approximately 20%, which is outside experimental error.

In summary, we have found the strong-coupling calculations for κ and H_{c2} near T_c to be in agreement with measurements on Pb-Bi- Tl and Pb-In alloys. Since both of these alloys derive their strong coupling from lead, it would be of interest to have an independent check from some nonlead,

- 'Research supported in part by the National Science Foundation under Grant No. Gu-2603.
- [†]Present address: Department of Physics, Slippery Rock State College, Slippery Rock, Pa. 16057.
- 'V. L. Ginzburgh and L. D. Landau, Zh. Eksp. Teor. Fiz. 20, 1064 (1950).
- 'L. P. Gorkov, Zh. Eksp. Teor. Fiz. 36, ¹⁹¹⁸ (1959) [Sov. Phys. JETP 9, 1364 (1959)]; Zh. Eksp. Teor. Fiz. 37, 833 (1960) [Sov. Phys. -JETP 10, 593 (1960)].
- ³K. Maki, Physics (N.Y.) 1, 21 (1964); C. Caroli, M. Cyrot, and P. G. de Gennes, Solid State Commun. 4, 17 (1966).
- 4G. Eilenberger, Phys. Rev. 153, 584 (1967).
-
- 5J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).
- ⁶g. Eilenberger and V. Amegaokar, Phys. Rev. 158, 332 (1967); Phys. Rev. B 2, 1433 (1970).
- 'K. D. Usadel, Phys. Rev. B 2, 135 (1970).
- ${}^{8}R$. C. Dynes and J. M. Rowell, Phys. Rev. 187, 821 (1969).
- ⁹D. E. Farrell, B. S. Chandrasekhar, and H. V. Culbert, Phys. Rev. 177, 694 (1969).

PHYSICAL REVIEW B VOLUME 8, NUMBER 1 1 JULY 1973

strong- coupling alloy.

ACKNOWLEDGMENTS

The authors wish to acknowledge many helpful discussions with R. L. Cappelletti, Lavern C. Clune, and J. F. Wagner who also prepared the Pb-In samples.

- ¹⁰J. G. Adler, J. E. Jackson, and T. A. Will, Phys. Lett. A 24, 407 (1967); J. G. Adler, J. E. Jackson, and B. S.
- Chandrasekhar, Phys. Rev. Lett. 16, 53 (1966).
¹¹CryoCal, Inc., Rivera Beach, Fla.
-
- 12 J. P. Frank and D. L. Martin, Can. J. Phys. 39, 1320 (1961).
- $13R$. W. Rollins and J. Silcox, Phys. Rev. 155, 404 (1967).
- ¹⁴W. A. Fietz, Rev. Sci. Instrum. 36, 1621 (1965). ¹⁵W. H. Kleiner, L. M. Roth, and S. H. Autler, Phys. Rev.
- 133, A1226 (1964).
- ¹⁶T. G. Berlincourt and R. R. Hake, Phys. Rev. 131, 140 (1963). ¹⁷F. W. Smith, A. Baratoff, and M. Cardona, Phys. Kondens.
- ¹⁷F. W. Smith, A. Baratoff, and M. Cardona, Phys. Kondens.
Mater. **12,** 145 (1970).
- ¹⁸D. L. Decker, D. E. Mapather, and R. W. Shaw, Phys. Rev. 112, 1888 (1958).
- ¹⁹R. F. Gasparovic, B. N. Taylor, and R. E. Eck, Solid State Commun. 4, 59 (1966).
- ²⁰H. V. Culbert, D. E. Farrell, and B. S. Chandrasekar, Phys. Rev. B 3, 794 (1971).
- 21 D. Johnson (private communication).

Thermomagnetic Effects in Dirty Transition-Metal Superconductors near the Upper Critical Field

Pramod Kumar and S. N. Gupta

Department of Physics, University of Roorkee, Roorkee, India

(Received 1 November 1972)

Using the Suhl, Matthias, and Walker two-band model, we have studied the thermomagnetic effects in dirty type-II transition-metal (TM) superconductors, in flux-flow state, immediately below the upper critical field and in the temperature region $T_c^{(0)}$ < $T < T_{cd} (= T_c)$. It is found that there is an anomalous increase of d-band thermomagnetic effects in dirty TM superconductors below H_{c2} , arising from the existence of interband impurity scattering. This behavior is analogous to the d -band Hall angle, recently investigated by Chow. For pure niobium, our results are exactly reducible to those of Caroli et al. for a one-d -band superconductor.

I. INTRODUCTION

In recent years, the Suhl, Matthias, and Walk $er¹$ (SMW) two-band model has been widely used to study and explain various physical properties $2-8$ of superconducting transition metals (TM) such as niobium and vanadium. In a recent publication, 6 we have successfully extended this model to study the thermal conductivity of dirty TM superconductors. These investigations, including that of Chow^{2,9} on specific heat, have confirmed that it is pair breaking as a result of interband impurity

scattering which is responsible for the anomalous changes in the various physical properties of these TM superconductors. It was assumed that the sband density of states is much smaller than the d band density of states at the Fermi energy. In all these investigations, it is assumed that the intraband BCS coupling constants g_s and g_d are nonzero, while the interband BCS coupling g_{sd} is assumed to be zero.

Very recently, Chow¹⁰ has investigated how the interband impurity scattering would influence the Hall effect of the dirty type-II TM superconductors