

## de Haas-van Alphen effect in dilute Pb-Tl and Pb-Bi alloys\*

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The changes in de Haas-van Alphen frequencies and Dingle temperatures in lead due to the addition of small amounts of thallium and bismuth impurities have been studied. The rigid-band model is obeyed at least to impurity concentrations of 0.2%. The effect of these impurities upon the Dingle temperature appears to be larger for the hole surface than the multiply connected electron surface.

### I. INTRODUCTION

The effects of impurities on carrier scattering and Fermi surfaces of metals have recently become an important topic of research. Because of advances in experimental techniques it is now possible to measure the effects of very small impurity concentrations, less than 0.1%, on the de Haas-van Alphen (dHvA) frequencies. In addition, by use of the dHvA effect it is possible to measure carrier lifetimes on specific known sections of the Fermi surface.

In the present paper we describe measurements of the dHvA effect in  $\text{Pb}_{1-C}\text{Tl}_C$ ,  $\text{Pb}_{1-C}\text{Bi}_C$ , and  $\text{Pb}_{1-C}\text{Tl}_{C/2}\text{Bi}_{C/2}$  alloys, for values of concentration  $C$  between 0 and about 0.005. This work supplements a previous paper on Pb-Bi alloys by Anderson and Hines,<sup>1</sup> which will be referred to subsequently as AH.

Our motivation to study lead alloys was threefold. First, a wealth of theoretical and experimental information had already been obtained for pure lead, but some ambiguities remained which could possibly be settled by varying the electron concentration. Second, since the Fermi surface is not too complicated,<sup>2</sup> it would be possible to separate the dHvA contributions from different parts of this surface. Finally, previous studies have shown that good pure-lead<sup>3</sup> and lead-alloy<sup>1</sup> crystals can be obtained by relatively straightforward sample-preparation techniques and that the effects of even small amounts of impurities should be observable.

The Fermi surface of lead consists of two pieces, a large second-zone closed hole surface centered at  $\Gamma$  and a third-zone multiply connected surface of arms in  $[110]$  directions.<sup>4</sup> We have studied (110) cross sections of both the hole surface, orbit  $\psi$ , and the arms, orbit  $\zeta$ , as well as a (100) cross section at the junction of the arms, orbit  $\nu$ .<sup>5</sup> These correspond to oscillations  $\alpha$ ,  $\gamma$ , and  $\beta$ , respectively.<sup>5</sup>

In Sec. II we describe our experimental tech-

niques including *in situ* NMR (nuclear-magnetic-resonance) measurements of magnetic field. In Sec. III the experimental results are presented, and these are compared with simple model-potential calculations in Sec. IV.

### II. EXPERIMENT

For our dHvA-effect studies we have used the conventional low-frequency field-modulation technique as described by AH with the dc magnetic field  $H_0$  produced by a 55-kOe superconducting solenoid. The NMR system for *in situ* magnetic field measurements has been improved so that magnetic field measurements are almost routine.<sup>6</sup> Because rf levels of less than 1-mV peak were possible with this system, we were able to use the <sup>23</sup>Na, <sup>133</sup>Cs, and <sup>35</sup>Cl resonances in NaCl and CsCl doped with NiCl<sub>2</sub> for our field measurements. The NMR linewidths for these specimens were less than 1 Oe and the field could easily be determined with an error of less than 10% of this width, if one neglected possible errors in the assumed NMR frequency-field relation. This frequency-field relation was determined at fixed magnetic fields by comparing the <sup>23</sup>Na, <sup>133</sup>Cs, and <sup>35</sup>Cl resonances with the <sup>27</sup>Al resonance in a metal powder (1.1112 MHz/kOe when corrected for the Knight shift).<sup>6</sup>

One complication arose because the same modulation was used for both the dHvA effect and the NMR calibration. At high magnetic fields the required field-modulation amplitude  $h_m$  might be as much as 50 Oe, and of course the advantage of a narrow NMR line was then lost. However, even under these circumstances it was possible to determine the center of the NMR line to better than 10% of the width, and therefore field measurements under the worst conditions had a precision of at least one part in 10<sup>4</sup>. Moreover, we found that it was possible to switch the field modulation from a high level, say 50 Oe, to a low level, say 1 Oe, at a rate of between 0.1 and 5 Hz. This "modulation" of the modulation took the form of a square

wave. By careful choice of this square-wave switching frequency, about 5% of the normal sinusoidal modulation frequency, it was possible to obtain reasonably satisfactory dHvA signals corresponding to the periods of large  $h_m$  and fairly narrow NMR lines corresponding to the periods of small  $h_m$ . Even though the NMR signal showed structure related to both amplitudes of modulation, the central portion of the resonance appeared sharply defined and the zero crossing could be determined quite reliably and precisely ( $\pm 0.2$  G at high fields).

There appeared to be two primary sources of error in our dilute-alloy studies: (i) inexact determination of sample orientation relative to the magnetic field, and (ii) contributions from more than one dHvA frequency to the signal. The former was the more important in our determination of dHvA frequencies.

In order to properly align a sample along a symmetry direction, it was necessary to be able to rotate the sample about two different axes independently. For this purpose we constructed a sample holder which allowed  $360^\circ$  rotation of the sample relative to a fixed vertical plane about a vertical axis and subsequent tilting from the vertical plane by up to  $5^\circ$ .<sup>7</sup> (This rotation system is similar to that described by Coleridge and Templeton.<sup>8</sup>)

The actual process of alignment was rather tedious, even though the required changes in angle were less than  $1^\circ$ . For some directions, particularly [100], it was possible to find the correct symmetry orientation by studying beats in the oscillation patterns. The  $\gamma$  branches cross at [100]<sup>4</sup>, and therefore the number of cycles per beat of the  $\gamma$ 's varies rapidly with angle, while the  $\beta$ 's, which were the oscillations studied at this orientation, have an extremum. From the beats in the  $\gamma$ 's we estimated an error in angle which was always less than  $0.4^\circ$ , corresponding to an error of less than 0.02% in the  $\beta$  dHvA frequency.<sup>9</sup> (In the most accurately aligned samples we estimate the error in dHvA frequency due to misorientation to be less than 0.003%.) Unfortunately, for the [110] direction it was actually necessary to adjust the orientation until the minimum dHvA frequency was found; this was not only more time consuming but also more difficult to verify, because of the beats in both the  $\gamma$  and  $\alpha$  oscillations at [110]. Thus the dominant error in our (110) dHvA-frequency measurements resulted from sample misalignment and in the worst case we estimate it to be about 0.04%. In more usual cases the errors were about 0.01%.

We should also mention here another complication in the  $\gamma$  oscillations for [110] orientation. The existence of beats of about 42.5 cycles for the magnetic field along [110] has been well established<sup>4</sup>;

however, in some samples the pattern was more complicated, consisting of a longer beat in addition to the 42.5-cycle beat, suggesting the presence of at least one more frequency. We believe that this additional frequency must result from occlusions of crystallites of slightly different orientation in some of our samples, because the number of cycles in the long beat varied from sample to sample, from a minimum of about 280 cycles to a number larger than we could measure. In some of our "best" samples there was no evidence for these long beats. We might also mention that the number of cycles per long beat did not appear to change for a variation in sample orientation of a few degrees. We have concluded that there are only two fundamental [110]  $\gamma$  frequencies, and additional frequencies should be attributed to sample preparation difficulties and not any fundamental aspect of the Fermi surface. Occlusions might also account for the observation of a third frequency in some Pb-In alloys, as reported by Tobin *et al.*<sup>10</sup>

The simplest satisfactory third-zone Fermi-surface model—that is, a model with only a maximum and minimum (110) arm cross-sectional area between  $U$  and  $W$ —would predict the largest cross-sectional area for the central section (through  $U$  or  $K$ ). From the early model of Anderson and Gold,<sup>4</sup> however, it was postulated that the two  $\gamma$  frequencies resulted from a minimum area at  $U$  and a point of inflection between  $U$  and  $W$  corresponding to neither a minimum nor maximum area. In our studies of carefully prepared pure-lead crystals we have established that the dominant oscillations correspond to the higher dHvA frequency and we believe that it is more likely that the dominant oscillations should result from a central section. (In principle it should be possible to check this point by a careful study of the angular dependence of the two  $\gamma$  frequencies near [110]. Such an experiment has not yet been carried out.) Therefore, we assume that the form of the third zone with only a maximum arm cross section at  $U$  (or  $K$ ) and minimum somewhere nearer  $W$ , and not the early model of Anderson and Gold,<sup>4</sup> is correct. This is in agreement with the calculations of Van Dyke.<sup>11</sup> (The nonlocal model-potential calculation of Anderson *et al.*<sup>9</sup> was not carried out with sufficient accuracy to check this point.)

The samples were prepared by the Czochralski technique in the same manner as described by AH. The pulled crystals were 2–3 mm in diameter and 5–10 cm in length. Samples of 0.5–1 cm in length were cut by the floating-acid technique.<sup>4,3</sup> The impurity (thallium, bismuth, or both for the iso-electronic alloys) was added to the melt and the resulting concentrations (nominal) were estimated from the concentration in the melt. Impurity

TABLE I. Tl-impurity concentration measurements in  $Pb_{1-c}Tl_c$ .

Sample orientation	Tl concentration (%)		
	Nominal	Measured	Variation
[100]	0.1	0.094	$\pm 0.015^a$
[110]	0.1	0.104	$\pm 0.016^a$
[100]	0.2	0.183	$\pm 0.043^b$
[110]	0.2	0.195	$\pm 0.029^a$
[100]	0.3	0.310	$\pm 0.046^a$
[110]	0.3	0.274	$\pm 0.028^b$

<sup>a</sup>Variation estimated to be  $\pm 15\%$ .

<sup>b</sup>Variation measure by spark-source mass spectrometer (Ref. 14).

analyses were made on six samples with Tl impurity by means of atomic absorption and the results are shown in Table I. As one can see from this table, the nominal values are quite close to the measured values. Since all of our samples were prepared in the same manner, we have assumed that nominal concentrations of Tl impurity are the actual concentrations for all samples for which the concentration has not been measured. This result is reasonable because the equilibrium distribution coefficient has been shown to be greater than  $1^{12,13}$  and under our pulling conditions the effective distribution coefficient should be reduced to about 1.

In contrast, as was discussed by AH, the equilib-

rium distribution coefficient for Bi in Pb is much less than 1, but it increased under our pulling conditions only to about 0.8. Consequently, the nominal and measured concentrations did not agree very well for bismuth impurity.

The uniformity of impurity concentration has been measured with a spark-source mass spectrometer for two of our  $Pb_{1-c}Tl_c$  samples (see Table I), and with a microprobe for two of our isoelectronic  $Pb_{1-c}Tl_{c/2}Bi_{c/2}$  samples.<sup>14</sup> (The microprobe was capable of looking at only the Bi impurity.) The over-all variation in impurity concentration was found to be about  $\pm 25\%$ ; however, the central portion of the sample, which is the portion normally studied, appeared to have a variation of concentration of less than  $\pm 10\%$ . Again, because all samples were prepared in the same manner, variations in impurity concentration should have been similar, and therefore we have taken a conservative estimate for all samples of  $\pm 15\%$ , which is the value used to estimate the variations shown in Table II.

Because of the differences in effective distribution coefficient one might expect less bismuth than thallium in the isoelectronic samples. The small negative value of  $\Delta F/F$  for the nominal 0.2% isoelectronic sample (Table II) may be evidence of this difference, but even here the effect is almost negligible.

TABLE II. Summary of alloy results.

C (nominal) (%)	Impurity	Approximate number of cycles	dHvA frequency $F$ (mG)	$\Delta F/F$ (%)	$X$ (°K)
	[110] $\alpha$	oscillations			
0		1800	159.15 $\pm$ 0.05	0 $\pm$ 0.04	0.4 $\pm$ 0.25
0.1	Tl	900	159.23 $\pm$ 0.03	0.05 $\pm$ 0.02	1.1 <sub>3</sub> <sup>+0.4</sup> <sub>-0.1</sub>
0.2	Tl	350	159.33 $\pm$ 0.03	0.11 $\pm$ 0.02	2.46 $\pm$ 0.18
	[110] $\gamma$	oscillations			
0		850	18.072 $\pm$ 0.004	0 $\pm$ 0.03	0.14 $\pm$ 0.05
0.1	Tl	700	18.034 $\pm$ 0.009	-0.21 $\pm$ 0.05	0.68 $\pm$ 0.03
0.2	Tl	500	18.005 $\pm$ 0.005	-0.36 $\pm$ 0.01	1.28 $\pm$ 0.05
0.1	Bi	750	18.086 $\pm$ 0.004	0.09 $\pm$ 0.01	0.4 <sub>5</sub> $\pm$ 0.2
0.5	Bi	200	18.14 <sub>5</sub> $\pm$ 0.02	0.4 $\pm$ 0.1	2.4 $\pm$ 0.3
	[100] $\beta$	oscillations			
0		1400	51.245 $\pm$ 0.002	0 $\pm$ 0.004	0.10 $\pm$ 0.01
0.1	Tl	600	51.204 $\pm$ 0.01	-0.08 $\pm$ 0.02	0.6 $\pm$ 0.1
0.2	Tl	350	51.108 $\pm$ 0.004	-0.267 $\pm$ 0.007	1.0 $\pm$ 0.2
0.3 <sup>a</sup>	Tl	200	51.02 <sub>7</sub> $\pm$ 0.02	-0.42 <sub>5</sub> $\pm$ 0.04	2.4 <sub>-1.0</sub> <sup>+0.2</sup>
0.3 <sup>b</sup>	Tl	200	51.00 $\pm$ 0.03	-0.48 <sub>5</sub> $\pm$ 0.06	2.8 <sub>-1.0</sub> <sup>+0.2</sup>
0.1 <sup>c</sup>	Tl+Bi	600	51.250 $\pm$ 0.009	0.009 $\pm$ 0.02	0.65 $\pm$ 0.15
0.1 <sup>c</sup>	Tl+Bi	600	51.251 $\pm$ 0.006	0.01 $\pm$ 0.01	0.70 $\pm$ 0.15
0.2 <sup>c</sup>	Tl+Bi	300	51.226 $\pm$ 0.009	-0.03 <sub>7</sub> $\pm$ 0.02	1.4 $\pm$ 0.2

<sup>a</sup>Measured concentration 0.31%.

<sup>b</sup>Measured concentration 0.274%.

<sup>c</sup>Isoelectronic alloys. Total nominal concentration is given.

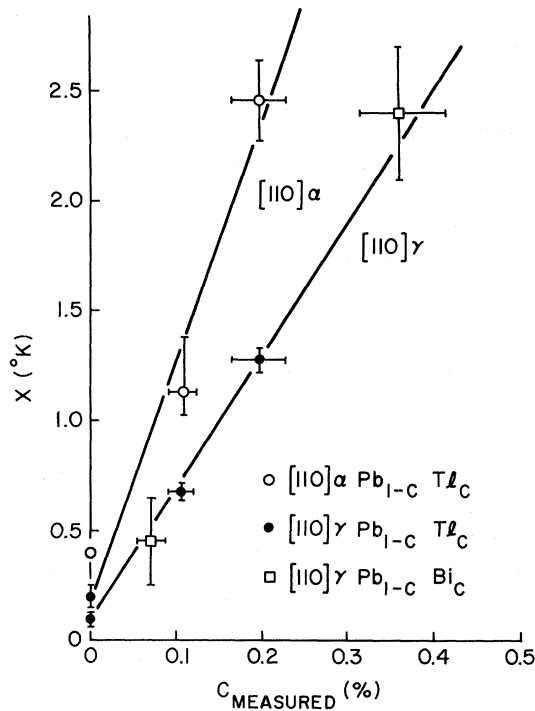


FIG. 1. Change in Dingle temperature vs measured concentration of thallium or bismuth impurity for the  $\alpha$  and  $\gamma$  oscillations.

### III. RESULTS

In Table II we list the samples that were investigated. In addition, many measurements were made on a number of other pure-lead crystals for which the results of Table II are representative. Since the number of measurable cycles decreased dramatically with increasing impurity concentration, these values are also given in Table II.

We have studied two [110] axis samples with nominal bismuth impurity concentrations of 0.1 and 0.5%. By comparison with the chemical analysis of similar Pb-Bi samples reported by AH, we have assumed that the ratio of actual concentration of bismuth to nominal concentration is 0.72 for the present samples. Further confidence in the correctness of this assignment may be obtained by noting in Fig. 1 that both Bi and Tl impurities have about the same effect on the Dingle temperature for the (110) oscillations.

#### A. Frequency changes

In Fig. 2 we present measurements of the relative changes of dHvA frequencies versus impurity concentration for both [110]  $\alpha$  and [110]  $\gamma$  oscillations due to addition of thallium. In addition, we show two results for the relative change in the [110]  $\gamma$  frequency for addition of bismuth impurity.

The lines correspond to the rigid-band-model (RBM) prediction, which takes the form

$$\frac{\Delta F}{F} = \left( \frac{m_0 c}{\hbar e} \right) \left( \frac{\mu}{\eta_0(E_F) F} \right) CZ. \quad (1)$$

Here  $F$  is the dHvA frequency,  $m_0$  the free-electron mass,  $c$  the velocity of light,  $\hbar$  Planck's constant divided by  $2\pi$ , and  $e$  the charge of the electron. The quantity  $\mu$  is the cyclotron mass ratio and we have used the experimental values of 1.10, 1.22, and 0.56 for the [110]  $\alpha$ , [100]  $\beta$ , and [110]  $\gamma$  oscillations, respectively.<sup>3</sup> The density of states for lead,  $\eta_0(E_F)$ , is  $7.93857 \times 10^{11} \text{ erg}^{-1} \text{ atom}^{-1}$  and was obtained from the electronic specific-heat coefficient.<sup>15</sup> The quantity  $Z$  is the difference in electron/atom ratio between lead and the impurity atom, while  $C$  is the relative concentration of impurity atoms.

We note that within experimental error both the [110]  $\alpha$  and [110]  $\gamma$  frequencies of the alloys with thallium impurity follow the rigid-band predictions. In addition, as expected, a positive shift for the  $\alpha$  oscillations and a negative shift for the  $\gamma$  oscil-

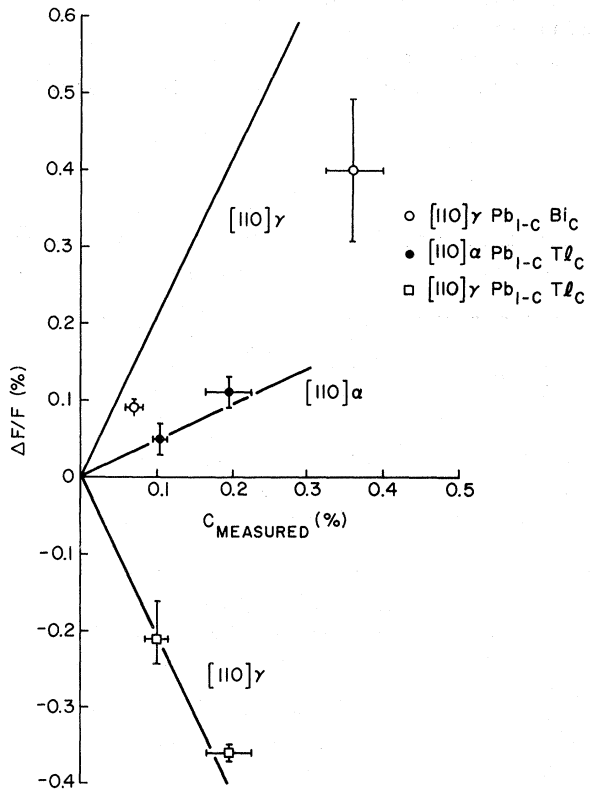


FIG. 2. Relative change in dHvA frequency vs measured impurity concentration for the  $\alpha$  and  $\gamma$  oscillations. Solid lines are RBM predictions using measured values of  $\mu$  and  $\eta_0(E_F)$ .

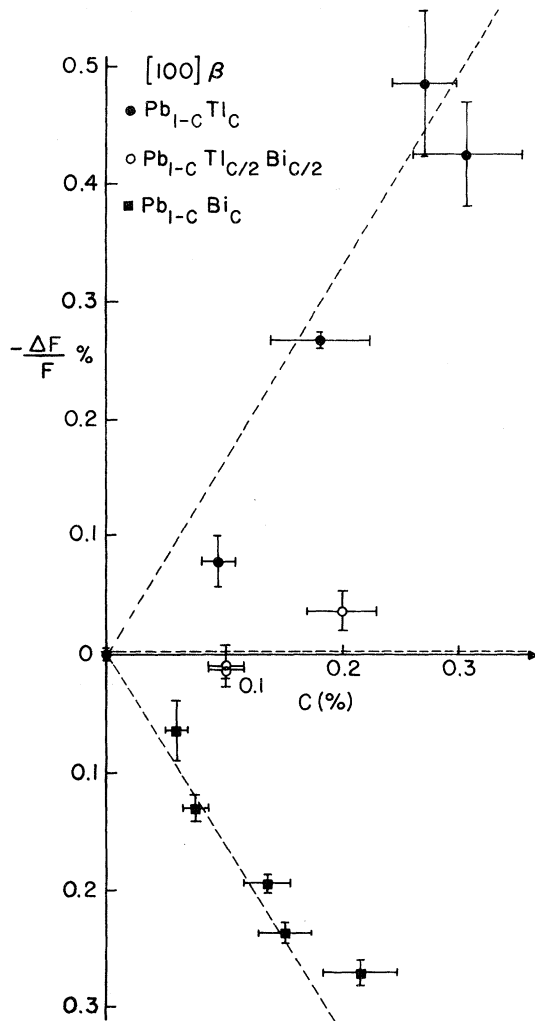


FIG. 3. Relative change in dHvA frequency as a function of impurity concentration for the  $\beta$  oscillations. Dashed lines are RBM predictions using measured values of  $\mu$  and  $n_0(E_F)$ .

lations confirm correspondence to a hole surface and an electron surface, respectively. On the other hand, the two measurements of the  $[110]$   $\gamma$  frequencies for bismuth impurity lie below the rigid-band prediction. Furthermore, for the nominal 0.5%-Bi sample approximately 200 dHvA cycles were observed and the 42.5-cycle beat seemed to have disappeared, indicating a change in Fermi-surface shape with alloying.

In Fig. 3 we show the relative change in frequency versus concentration for the  $[100]$   $\beta$  oscillations. In addition to the results for Tl impurity and for the isoelectronic case with equal amounts of Tl and Bi, we present for completeness the Bi impurity results of AH. The fact that the frequency decreases for Tl impurity, which has one less valence electron than Pb, provides further

confirmation that the  $\beta$  oscillations are produced by an electron surface. The data agree quite well with the RBM, which is shown by the straight lines.

For the isoelectronic alloys the dHvA frequencies are almost unchanged, which shows that the five-valence-impurity Bi almost exactly compensates the three-valence-impurity Tl.

#### B. Dingle temperatures

Dingle temperatures were obtained from the relationship given by

$$\frac{-\lambda\mu}{H_0} X = \ln[AT^{-1}H_0^{1/2} \sinh(\lambda\mu T/H_0)] + \text{const}, \quad (2)$$

where  $A$  is the amplitude of the dHvA oscillations,  $\lambda = 147 \text{ kOe}/^\circ\text{K}$ , and  $H_0$  is the applied quasistatic magnetic field.

Because the modulation amplitude was varied as  $H_0^2$ , the Bessel functions, occurring in the usual expression for field modulation experiments, was a constant and could be included in  $A$ . Thus a plot of the right-hand side of Eq. (2) versus  $-\lambda\mu/H_0$  is expected to yield a straight line of slope  $X$ . However, because of the difficulty in isolating each particular dHvA frequency, this simple approach was not always completely successful.

The complication of more than one dHvA frequency was particularly serious for the  $[100]$   $\beta$  oscillations, where there were two frequencies differing by about 0.17%. This resulted in beats of roughly 570 cycles and produced the greatest uncertainty in the determination of  $X$ . For pure samples for which more than 1400 cycles could be observed it was possible to measure amplitudes of corresponding portions of each beat cycle and thus obtain a definite slope for the Dingle-temperature plot from the beat envelope. For the 0.1%-impurity samples approximately one complete beat cycle was observed and could be corrected for in the determination of the Dingle temperature. However, for larger impurity concentrations less than one complete beat cycle could be measured. In these cases the effect of beating was corrected for in determining the true amplitude of the oscillations by assuming the same beat pattern as for the 0.1% alloys, but the resulting errors were quite serious as shown in Table II.

For the  $[110]$   $\gamma$  oscillations the effects of the 42.5-cycle beats were not important, since many beat cycles could be observed even in the most impure samples investigated. On the other hand, for the  $[110]$   $\alpha$  oscillations there were long-period beats of about 350 cycles, probably due to beating between the  $\alpha$  oscillations and a mixing frequency corresponding to the sum or difference between the  $\alpha$  frequency and a frequency resulting from the difference between the  $\gamma$  frequencies mentioned

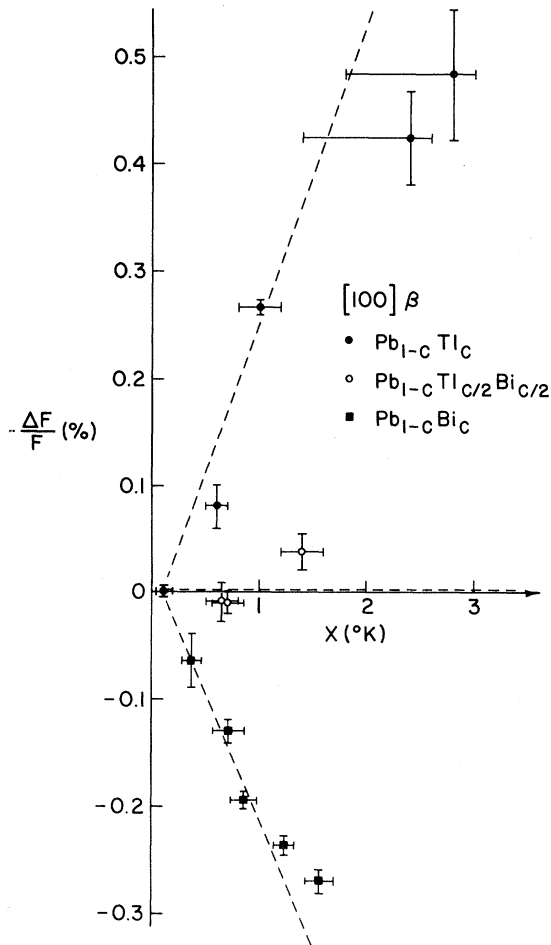


FIG. 4. Relative change in dHvA frequency vs Dingle temperature for the  $\beta$  oscillations. Dashed lines are RBM predictions using measured values of  $\mu$  and  $n_0(E_F)$ .

previously. This mixing frequency was probably a result of magnetic interactions, since its amplitude decreased drastically with impurity concentration and therefore only in the pure samples did this beating produce significant errors in the determination of  $X$ .

In Fig. 1 we show Dingle temperatures for the  $[110]$   $\gamma$  and  $[110]$   $\alpha$  oscillations. It is apparent that the change in  $X$  with concentration for the  $\alpha$  oscillations is nearly twice that for the  $\gamma$  oscillations. The straight lines represent least-squares fits to the data. For the  $\gamma$  oscillations, omitting the values obtained from samples with Bi impurity, we find for the Dingle temperature

$$X^\gamma = 0.09 + 592C . \quad (3)$$

We see that within experimental error the Bi-impurity samples also lie on the line of Eq. (3). For the  $\alpha$  oscillations the relationship between Dingle temperature and concentration is

$$X^\alpha = 0.29 + 1036C . \quad (4)$$

In Fig. 4, following the procedure of AH, we have plotted the relative frequency shifts versus Dingle temperature for the  $[100]$   $\beta$  oscillations because we believe the Dingle temperatures give a better representation of the concentrations of interest than chemical analysis. Of course, in order to obtain the lines corresponding to the rigid-band model it was necessary to determine the relationship between Dingle temperature and concentration for the  $\beta$  oscillations. For Tl impurity we obtained from a least-squares fit

$$X_{Tl}^\beta = 0.10 + 553C . \quad (5)$$

In making this fit we have, in effect, omitted the 0.3%-impurity samples because of the large errors associated with measurements on them. The thallium results agree within experimental error (about 15%) with the equation for Bi impurities given by AH:

$$X_{Bi}^\beta = 0.09 + 674C . \quad (6)$$

We have also plotted on the same graph the results for the isoelectronic samples by assuming that the scattering by the two impurities is additive.

#### IV. DISCUSSION

The measurements of relative frequency shifts due to either Bi or Tl impurities for both the hole and electron surfaces can be explained by the rigid-band model within experimental error, except possibly for the nominal 0.5%-Bi sample. Measurements of deviations from the RBM might permit a determination of the variation of density of states with energy at the Fermi level. We are still unable to state definitely whether the fourth zone is occupied or not in pure lead, although existing measurements plus nonlocal-model-potential calculations<sup>9</sup> suggest that the fourth zone lies several meV above the Fermi level.

The measurements with both Bi and Tl impurities show very conclusively that the  $\alpha$  oscillations originate from a hole surface, while the  $\beta$  and  $\gamma$  oscillations result from an electron surface.

Possibly the most interesting aspect of these results is the Dingle-temperature measurement. First we see that the variation of the Dingle temperature with impurity concentration is essentially the same for both bismuth and thallium impurities. In addition, as one might expect, the scattering appears to be additive for the two impurities in the isoelectronic alloys.

We have made a simple calculation to estimate the contribution by substitutional impurities to electron scattering. The total scattering probability is calculated by applying the Golden Rule;

the perturbation is the difference in Heine-Abarenkov model potential between lead,  $W^{\text{Pb}}$ , and the impurity  $W^i$ .<sup>16</sup> The total scattering probability is an integral over wave number  $q$ ,<sup>17</sup> given by

$$\sum_{k'} P_{kk'} = \frac{\Omega_0 m}{2\pi \hbar^3 N k} \int_0^{2k} |\langle \vec{k} + \vec{q} | W^i | \vec{k} \rangle - \langle k+q | W^{\text{Pb}} | \vec{k} \rangle|^2 q dq. \quad (7)$$

Here the atomic volume for Pb is  $\Omega_0$ ,  $m$  is the electron mass,  $N$  is the number of atoms in the crystal, and both initial and final states  $\vec{k}$  and  $\vec{k} + \vec{q}$ , respectively, lie on the Fermi surface. Effects of spin-orbit coupling have been neglected.

For a spherical Fermi surface a simple numerical calculation gives for the average collision time  $\tau$  for thallium and bismuth impurities, respectively,

$$\left(\frac{1}{\tau}\right)_{\text{Tl}} = \sum_{k'} P_{kk'} = 0.43 \times 10^{15} \text{ C sec}^{-1}, \quad (8)$$

$$(1/\tau)_{\text{Bi}} = 0.20 \times 10^{15} \text{ C sec}^{-1}.$$

As a result the variations in Dingle temperature with impurity become for Bi and Tl, respectively,

$$X_{\text{Tl}} = 523 \text{ C}^\circ \text{K}, \quad (9)$$

$$X_{\text{Bi}} = 243 \text{ C}^\circ \text{K}.$$

The value for Tl of 523 is in good agreement with the experimental value of 553 [Eq. (5)], but the value of 243 for Bi differs by nearly a factor of 3

from our measured value of 674 [Eq. (6)]. The discrepancy for Bi is probably indicative of the limitations of the model potential, especially near the core. Because the Bi ion has a charge of +5 compared to +3 for Tl, electrons are attracted more strongly to the Bi core and the potential near this core is more effective in scattering electrons than the potential near the core of Tl, but this part of the potential is not well represented by the model potential. This effect has also been noted by Tripp and Farrell.<sup>18</sup>

We also see from a comparison of the Dingle temperatures for the [110]  $\gamma$  and  $\alpha$  oscillations (Fig. 1) that the impurities are nearly twice as effective in scattering from the hole surface as from the electron surface. A useful approach for investigation of this scattering might be a parameterized phase-shift analysis following the Korringa-Kohn-Rostoker formalism,<sup>19</sup> but to our knowledge no such calculations have been made for simple metals and their alloys.

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