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Magnetic interaction in the de Haas–van Alphen effect in lead*

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The frequency modulation of the high-frequency de Haas–van Alphen oscillation from the second-band hole sheet of the Fermi surface with the period of the low-frequency oscillation from the third-band electron network along the [110] crystalline direction in Pb has been examined in ellipsoidal samples. Within the framework of the Shoenberg conjecture that the magnetic induction is the relevant field in the Lifshitz-Kosevich theory and the assumption that the induction is uniform over the sample, the analysis of the frequency modulation should yield an absolute determination of the magnetic induction oscillation amplitude. This determination in each of our samples was found to be inconsistent for two crystallographically equivalent directions. The results can most easily be interpreted as implying that the induction averaged over the cyclotron orbit has an oscillation amplitude about 25% larger than that averaged over the sample. Several mechanisms which could cause an inhomogeneous magnetic induction are considered.

I. INTRODUCTION

Lifshitz and Kosevich[1] (LK), building on earlier work by Landau,[2] Peierls,[3] and Onsager,[4] developed the detailed theory for the quantum oscillations in the induced magnetization of the noninteracting electron gas. This theory for what is known as the de Haas–van Alphen[5] (dHvA) effect is quantitatively accurate in most cases. Shoenberg[6] first observed deviations from the predictions of this theory, and he attributed these deviations to magnetic interaction among the electrons. He observed that the harmonic content of the strong belly oscillation in the noble metals became abnormally large and that the amplitude of the fundamental oscillation tended to saturate as the oscillation amplitude was increased either by increasing the applied magnetic field or reducing the temperature. Such effects would be obtained in the LK theory if the applied field, the only field in the noninteracting case, were replaced with a field containing a sample magnetization contribution; that is, if

$$B_r = H + 4\pi b' M , \qquad (1)$$

where B_r replaces the applied field H, at least in the argument of the oscillations in the LK theory. Here M is the magnetization, and b' is a strength parameter. Shoenberg originally proposed the explanation of his results by setting B_r equal to the macroscopic induction inside the sample; i.e., by setting $b' = 1 - \eta$, where η is the demagnetizing coefficient that ranges from zero for the infinite rod to 1 for the infinite disk. This proposal has become known as the Shoenberg conjecture (SC). There have been several theoretical justifications that the field appropriate for the dHvA effect is the magnetic induction. Pippard[7] has given a very direct thermodynamic argument supporting it. A justification within mean-field theory is given in a paper by Condon,[8] and it has been justified within the Hartree approximation by Kaplan and Glasser[9] and by Holstein, Norton, and Pincus.[10]

While it is clear that the arguments of the dHvA oscillations contain the oscillations themselves,[11-14] there has been as yet no experiment precise enough to quantitatively verify that B_r is the macroscopically averaged magnetic induction. This lack of needed precision has been caused primarily by the difficulties in evaluating the absolute values of the dHvA amplitudes as well as the precision that is needed to measure the relatively weak effects of magnetic interaction. In fact, there have been some indications that B_r may be substantially different from the sample-averaged induction.[15] The present experiment was designed to make a more precise evaluation of this effect. A direct analysis of our results seems to show an apparent disagreement with the SC taken in its narrow sense, i.e., with $b' = 1 - \eta$. We have adopted then an empirical formalism by writing $b' = b - \eta$ and treating b as a disposable parameter. Our results give consistent values for b which are substantially greater than 1. This led us to consider the possibility that the magnetization varies over the sample on a scale on the order of or larger than the cyclotron orbits of the electrons producing the magnetic interaction. Such a possibility could produce an induction averaged over a cyclotron orbit which, owing to dephasing, contains larger magnetization oscillations than the induction uniformly averaged over the volume of the sample and appropriate for the demagnetizing field. This is discussed in some detail in Sec. IV.

II. EXPERIMENT

Along the [110] crystalline direction in Pb there are two oscillations called γ that have frequencies of approximately 18×10^6 G and form a beat pattern of some 42.8 oscillations. These oscillations are now attributed to maximum and minimum cross sections on the third-zone electron network of the Pb Fermi surface.[16] Along this direction there is one other known dHvA oscillation called α with a frequency of 158.4×10^6 G that is attributed to the central orbit around the second-zone hole sheet.[17] With the applied magnetic field on the order of 51 kG and the sample temperature on the order of 1 K, the amplitudes of the γ oscillations are large enough to cause substantial frequency modulation of the α oscillation around the antinode of the γ beat pattern. Since dHvA magnetometers are set up to make very accurate frequency measurements, we have chosen to make our primary measurements on this frequency modulation.

Within this general approach the degree of modulation of the α frequency is determined by the product of b' with the net γ absolute amplitude, and measurement of the frequency modulation will evaluate this product. In order to determine b' then one must measure the absolute γ amplitude. The direct measurement of absolute dHvA amplitudes is very difficult and generally cannot be done to better than 10% or so. Since this is not sufficient for this work, we have opted for an indirect measurement. To ensure that the demagnetizing field is uniform we have prepared ellipsoidal samples. The samples were cut so that two $\langle 110\rangle$ crystalline directions lay along the longest and shortest of the ellipsoid axes. The demagnetizing coefficients along these two directions were calculated from the sample dimensions. Thus, by measuring the frequency modulation along the two crystallographically equivalent directions which have different, known η, a determination of both the γ amplitude and b can be made.

Four samples were cut with an acid erosion string saw from two different boules of Cominco 69 grade zone-refined lead which were purchased about four years apart. The first sample was a rectangular slab, and the data taken with this sample have been designated as from sample 1. The sample was then etched into a hemiellipsoid; i.e., one surface of the sample was rounded off so that the sample shape reasonably well approximated half of an ellipsoid. The data taken on this sample after etching have been designated as from sample 1A. Three other samples, designated samples 2, 3, and 4, were also cut with the acid saw. The shapes of these samples originally approximated those of elliptical slabs and were cut by making 20 or so straight-line cuts. The slabs were then etched on both sides into quite good ellipsoidal shapes. The etching was accomplished by placing a small bit of Kleenex on the place to be etched and adding a drop of the lead etching solution.[18] The outlines of each sample were traced on a traveling microscope after the data were taken. The deviation from true ellipsoidal shape was nearly the same for samples 2–4, and the outlines of sample 3 are shown in Fig. 1. The dimensions and demagnetizing factors for the four samples are given in Table I. The uncertainties associated with the demagnetizing factors are estimated from the irregularity of the sample shapes in the cases of samples 2–4.[19] The etched samples were placed on a quartz plate and inside a Vycor tube. The tube was well flushed and filled with a helium atmosphere which was always maintained at an overpressure. The tube was placed in a furnace, and the samples were annealed for three days at 320 °C. The entire system had previously been thoroughly cleaned and baked out under vacuum to avoid contamination of the samples.

The annealed samples were placed on a post whose end surface was cut at an angle of 45° with respect to the length of the post. This end of the post was covered with a thin film of silicone vacuum grease. The sample was picked up off the quartz plate by the adhesion of the grease for that small portion of the sample surface touching the post. The post was placed in a coil form at the center of a pickup coil of approximately 6000 turns of 46-gauge copper wire. The pickup coil was connected in series to a similar coil on the same form wound in opposition. The coils had been previously adjusted to have the same area turns to within $\frac{1}{8}$ of an outside turn. The geometrical ar-

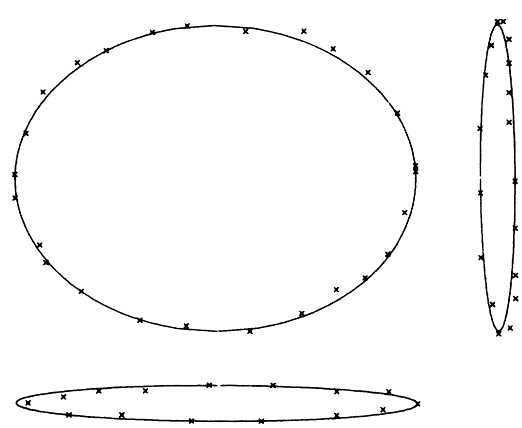

FIG. 1. Principal cross-section outlines of sample 3 as measured with a traveling microscope. The solid lines are the extremal cross sections of an ellipsoid with the dimensions given in Table I. The least count of the measurements is ±0.01 mm.

TABLE I. List of the ellipsoid-shaped lead samples with their principal dimensions. The longest and shortest sizes correspond to the [110] and [1$\bar{1}$0] directions, respectively, and the corresponding demagnetizing factors in these directions are given in column 3. Frequency modulation is indicated in columns 4 and 5 with the maximum and minimum values of the quantity f_α/f_α^*, with f_α the averaged frequency and f_α^* the apparent (modulated) frequency. The apparent Shoenberg factor a_γ and the empirical parameter b are determined by the methods indicated in the text. This a factor roughly corresponds to $4\pi dM/dH$ uniformly averaged over the volume of the sample, and that corresponding to a cyclotron orbit would be to a first approximation b times the quantity listed. Thus the "local" a factors were about 0.3 for the small demagnetizing direction.

Sample	Sample dimensions (ellipsoid axes) (mm)	Demagnetizing factors along the two ⟨110⟩ directions	$\frac{f_\alpha}{f_\alpha^*}\Big\vert_{max}$	$\frac{f_\alpha}{f_\alpha^*}\Big\vert_{min}$	Determined from Eq. (7) a_γ	b	Determined from Eq. (8) b_{min}	b_{max}	Weighted average
1-1A	4.95×3.20×0.62	0.073±0.020 0.791±0.020	1.352±0.014 1.127±0.011	0.779±0.006 0.895±0.008	0.237±0.020	1.28±0.05	1.20±0.07	1.44±0.11	1.30±0.09
2	6.90×5.20×0.75	0.068±0.010 0.828±0.010	1.340±0.014 1.123±0.008	0.777±0.007 0.915±0.004	0.233±0.016	1.27±0.08	1.26±0.06	1.30±0.05	1.28±0.01
3	6.90×5.20×0.60	0.056±0.010 0.859±0.010	1.353±0.011 1.114±0.004	0.768±0.004 0.922±0.002	0.245±0.012	1.25±0.05	1.24±0.03	1.27±0.03	1.26±0.01
4	7.00×1.75×1.06	0.050±0.010 0.592±0.010	1.393±0.010 1.225±0.006	0.734±0.003 0.860±0.003	0.271±0.015	1.26±0.06	1.32±0.07	1.19±0.04	1.24±0.04

rangement is shown in Fig. 2. The coil form was then placed in a jig made for the purpose, and the sample was x rayed. The coil and sample were adjusted so that ⟨100⟩ symmetry axes lay along the x-ray beam and the rotation axis of the sample holder into which the coil form was placed. This was the only time at which the samples were touched after they left the annealing furnace to give them a slight rotation on the post surface to perfect the orientation. The rotating sample holder has been described previously.[20] Thus two ⟨110⟩ directions lay 45° from the coil axis and in the plane of sample rotation. This orientation was done to within $\frac{1}{4}$°. Similar x rays were taken after each run to ensure that the sample had not lost its orientation. The coil form and sample were then placed in the sample holder and then into the Dewar in the bore of a 57-kG Westinghouse superconducting solenoid which was at room temperature. The sample was cooled to 55 K over a period of two to three hours. The large thermal capacity of the magnet ensured that the sample was cooled slowly and uniformly. The silicone vacuum grease only touched the sample over a small portion of its surface. Also the grease hardens below 100 K. All this considered, any thermally induced strains in the samples should have been minimal. The liquid helium was transferred first into the magnet space over a period of about an hour and a half, ensuring that the cooling of the sample from 55 to 4.2 K would also proceed slowly. A second helium bath was then transferred into the sample Dewar, and this bath was pumped to about 1 K. The rotating sample holder had been previously well calibrated, and the two ⟨110⟩ axes were indeed found to be 90° apart, as was determined from continuous field-rotation patterns. In addition, field sweeps at the two axes showed from the field position of the node

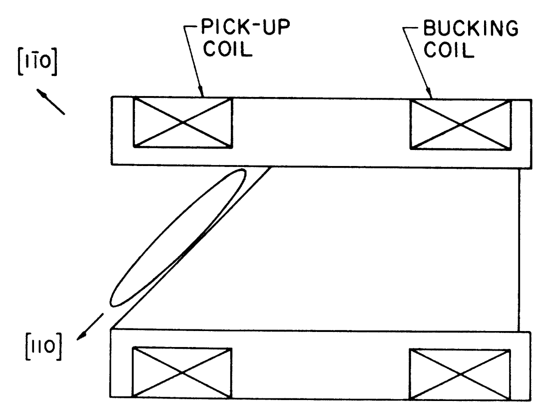

FIG. 2. Geometrical arrangement of the pickup and bucking coils relative to the sample. The crystalline direction [110] lies along the coil axis.

of the γ beat pattern that the "determined symmetry directions" were within $\frac{1}{4}°$ of the same crystallographic direction. The measured γ amplitudes at the two orientations were the same to within 3 or 4%, which is as good as one can expect for the possibly slightly different magnetization-vector-coil coupling in the two cases even with perfect crystallographic alignment with respect to the applied field.

A block diagram of the detection system is shown in Fig. 3. The magnetic field was swept inversely with time so that analog filtering of the detected signal could be accomplished. The amplitude of the modulation field ranged from 0.5 to 4.5 G, but a modulation amplitude of 1 G was used most of the time. Thus, the detection system was tuned to make the α oscillation dominant in the detected signal.[21] A small trace of the γ oscillation remained, and the output of the lock-in amplifier was passed to a Krohn-Hite model 3342 filter to remove the remaining γ oscillation. We also took a fair amount of data with no filter at all to ensure that the filter was not introducing spurious results. The measurement of the frequency modulation was not much affected by the small amount of remaining γ oscillation so reasonable measurements of the frequency modulation could be made without filtering. These results were consistent with those of the filtered data. The sweep rate was such that the average time frequency of the α oscillation was approximately 0.6 Hz. The signal was recorded on a Hewlett-Packard model 7100 B strip-chart recorder. After the field-rotation patterns were made to locate the ⟨110⟩ axes, approximately 15 field sweeps were made, alternately at both axes for each sample, by rotating *in situ* back and forth. The frequencies were determined from the chart position of alternate zero crossings of the oscillations. The field values were determined by doing

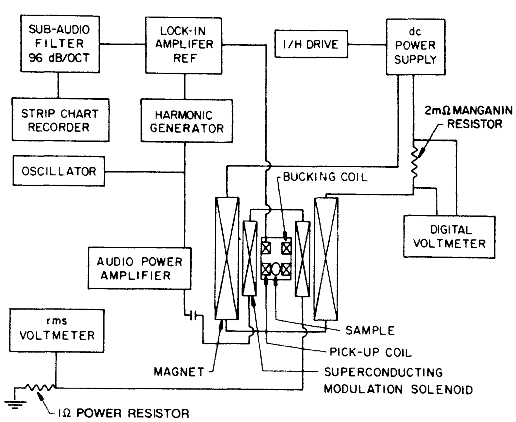

FIG. 3. Block diagram of the detection system.

a least-squares fit for each sweep of chart position to inverse field from 10 or 12 field markings.[22] The sweep was known to be highly $1/H$.[23] While it was possible for the absolute field values to have been offset slightly due to trapped flux, the differences between field values should have been quite accurate.[24]

Certain experimental artifacts that can directly affect the measurements are known to be induced when the dHvA amplitudes become large enough to cause observable magnetic interaction. Because we were concerned with the possible importance of these problems, we have examined them in some detail. Magnetic interaction itself results in the creation of sum and difference frequencies, or sidebands, which produce a modulation of the α frequency with the period of the γ frequency but do not cause an amplitude modulation in the magnetization itself. However, when the field-modulation method is used, the detected signal consists of a sum of derivatives of the magnetization which, owing to the frequency modulation, does contain an amplitude modulation. This FM-AM effect which has been discussed by Alles and Lowndes[14] causes most of the amplitude modulation of the α oscillation which has been observed but does not affect significantly the measurement of the frequency modulation. Plummer and Gordon[25] have shown that in the presence of a strong dHvA oscillation eddy currents can cause an oscillating skin depth for the modulation field. This causes the various portions of the sample to contribute to the detected signal with a sensitivity that contains a slight oscillation and produces an amplitude modulation of the high-frequency signal.[26] While this should not affect the frequency modulation, we were concerned about the potential problems and varied as many of the experimental parameters as may have an effect. We used modulation frequencies of 200, 100, 50, 20, and 10 Hz. We detected at the fundamental frequency and the second and fourth harmonic of it. As mentioned above we varied the amplitude of the modulation field by nearly an order of magnitude. The only effect we observed was the expected variation of the amplitude modulation. Plummer and Gordon have also shown that when eddy currents become effective a phase roll of the detected signal with respect to the modulation field with the period of the strong dHvA oscillation occurs. We looked for this phase roll and did not observe it, indicating as well that eddy currents were not a substantial problem.[27] Hornfeldt, Ketterson, and Windmiller,[28] have shown that an inhomogeneous applied magnetic field can cause an oscillatory phase smearing of the high-frequency oscillation over the sample and produce an amplitude modulation of the high-frequency oscillation with the period of the strong low-frequency oscillation, without noticeably affecting the frequency modulation. The field profile supplied with the magnet showed that the homogeneity was such that the field did not vary by more than 1 G over the sample. Under the approximations used by Hornfeldt, Ketterson, and Windmiller[29] this inhomogeneity should not introduce any measurable amplitude modulation. To ensure that indeed there was no effect on the frequency modulation we raised and lowered the sample 6 mm from the homogeneous portion of the field in increments of 1.5 mm. Although the overall amplitude of the signal was affected by the inhomogeneity, no noticeable effect on the frequency modulation was observed.[30]

III. RESULTS

In all more than 100 field sweeps for the four samples have been reduced. This large number was necessary in order to statistically reduce the uncertainty in the frequency values which are determined. The quantities of interest are the maximum and minimum α frequencies which occur at π and 0 phase of the γ oscillation, respectively. These results are listed in Table I as a ratio of the unperturbed α frequency to the maximum and minimum values of the modulated frequency. The uncertainties listed are the usual standard deviations of the mean for each series of measurements. Generally speaking samples 3 and 4 yielded the best data. Sample 1-1A was not a good ellipsoid in either case, and these being our earliest data the signal-to-noise ratio was higher than it was with the succeeding data. During part of the run on sample 2 a steel stool was inadvertently left near the magnet. Since it was possible for the stool to have disturbed the field homogeneity we have not spent the time to reduce as much of the data for this sample as we have for the others. However, the results for this sample are fully consistent with those for the others. Representative field sweeps for sample 3 along the small and large demagnetizing directions are shown in Fig. 4. The assignment of the amplitude modulation observed in Fig. 4 to various sources is somewhat complex owing to the phase differences among the various contributions. The contribution of the FM-AM effect is straightforward to determine and accounts for most of the observed amplitude modulation. The contribution from the eddy-current-induced oscillating skin depth has been estimated to have a value of about 15% of the FM-AM contribution.[31] Contributions from other detection-scheme artifacts such as field inhomogeneity have been determined to be negligible in this experimental arrangement. These two contributions from the FM-AM effect and the eddy currents reproduce the observed amplitude modulation to within 10%, which is about as good agreement as one could expect. A third possible contribution to the amplitude modulation, phase smearing, is discussed in Sec. IV. The frequency modulation determined from the sweep shown in Fig. 4(b) is shown in Fig. 5.

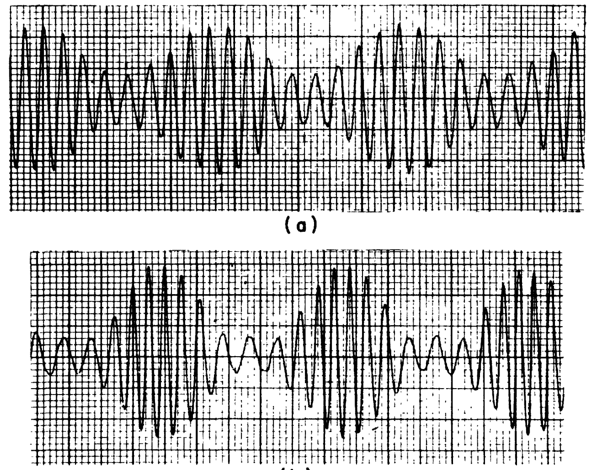

FIG. 4. Detected signal for sample 3 with the field direction along (a) the large demagnetizing direction and (b) the small demagnetizing direction. For both of these sweeps: modulation 1.94-G amplitude at 100 Hz, detection at second harmonic, filtering at 48 dB/octave, high pass.

If the two γ oscillations are treated as a single oscillation, the sample magnetization is of the

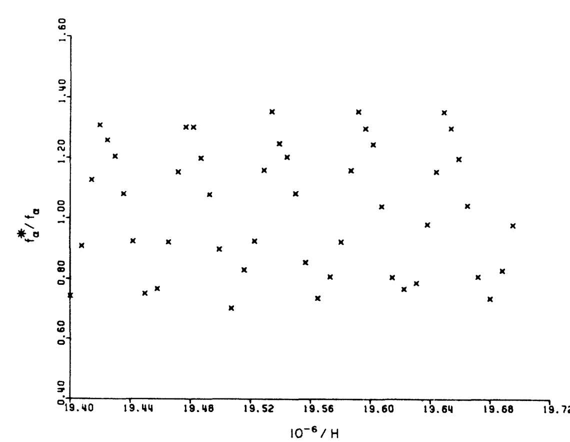

FIG. 5. Frequency modulation determined from the sweep of which a portion is shown in Fig. 4(b). The uncertainty in the determination of the frequency modulation was reduced substantially from that corresponding to a single sweep by averaging many such sweeps.

form

$$M = M_\alpha + M_\gamma = \sum_{j=1}^{\infty} M_{j\alpha} \sin(j\phi_\alpha + \delta_{j\alpha}) + \sum_{j=1}^{\infty} M_{j\gamma} \sin(j\phi_\gamma + \delta_{j\gamma}) . \quad (2)$$

The phases are given by

$$\phi_\alpha = 2\pi f_\alpha B_r^{-1} = \frac{ch}{e} S_\alpha B_r^{-1} ,$$
$$\phi_\gamma = 2\pi f_\gamma B_r^{-1} = \frac{ch}{e} S_\gamma B_r^{-1} , \quad (3)$$

where S_α and S_γ are the Fermi-surface cross sections corresponding to the electron orbits that produce the magnetization oscillations. The dHvA frequency then appears as

$$f = \frac{1}{2\pi} \frac{d\phi}{dB_r^{-1}} .$$

However, since the oscillations are recorded as a function of the applied field, the apparent frequency for the α oscillation is then

$$f_\alpha^* = \frac{1}{2\pi} \frac{d\phi_\alpha}{dH^{-1}} = f_\alpha \frac{dB_r^{-1}}{dH^{-1}} \simeq f_\alpha \frac{dB_r}{dH} , \quad (4)$$

which oscillates with the period of the strong oscillations since dB_r/dH oscillates. Then

$$\frac{f_\alpha}{f_\alpha^*} \simeq \frac{dH}{dB_r} = 1 + a_\gamma b' \sum_{j=1}^{\infty} j \frac{M_{j\gamma}}{M_{1\gamma}} \cos(j\phi_\gamma + \delta_{j\gamma}) + a_\alpha b' \sum_{j=1}^{\infty} j \frac{M_{j\alpha}}{M_{1\alpha}} \cos(j\phi_\alpha + \delta_{j\alpha}) , \quad (5)$$

where

$$a_\gamma = 8\pi^2 f_\gamma M_{1\gamma}/B_r^2$$

and

$$a_\alpha = 8\pi^2 f_\alpha M_{1\alpha}/B_r^2 ,$$

which are the usual Shoenberg "a factors." We can obtain a particularly simple and usable expression for $a'_\gamma = b' a_\gamma$ by keeping only the first two terms in the harmonic expansion for M_γ. The amplitudes of the third- and higher-harmonic terms are sufficiently small that their neglect will not introduce serious error.[32] The α magnetization is small enough that it can also be neglected.[33] The ratio of the true to the apparent frequency then oscillates between the extremes

$$\left.\frac{f_\alpha}{f_\alpha^*}\right|_{\max} \simeq 1 + a'_\gamma + \epsilon ,$$
$$\left.\frac{f_\alpha}{f_\alpha^*}\right|_{\min} \simeq 1 - a'_\gamma + \epsilon , \quad (6)$$

where

$$\epsilon = 2a_\gamma b' (M_{2\gamma}/M_{1\gamma}) \cos(\delta_{2\gamma} - 2\delta_{1\gamma}) .$$

The second-harmonic term cancels if we take the difference, and

$$a'_\gamma \simeq \frac{1}{2}\left(\left.\frac{f_\alpha}{f_\alpha^*}\right|_{\max} - \left.\frac{f_\alpha}{f_\alpha^*}\right|_{\min} \right) . \quad (7)$$

A value for a'_γ can be determined from the maximum and minimum values of the frequency ratio for both orientations in each sample. If the two orientations are indicated by subscripts 1 and 2, the two values $a'_{\gamma 1} = b'_1 a_\gamma$ and $a'_{\gamma 2} = b'_2 a_\gamma$ together with the known values of η_1 and η_2 for the corresponding demagnetizing factors are not consistent with $b' = 1 - \eta$. This unexpected behavior led to the adoption of the empirical formalism with $b' = b - \eta$. Then the two values of $a'_{\gamma 1} = (b - \eta_1) a_\gamma$ and $a'_{\gamma 2} = (b - \eta_2) a_\gamma$ can be solved for a_γ and b.

The harmonic content of the γ oscillation is sufficiently small that its neglect should not introduce any serious error into the determination of b and a_γ. However, Eq. (5) can be solved for b, although not a_γ, including all of the γ harmonics and neglecting only the α oscillation. Although the corresponding $(f_\alpha/f_\alpha^*)_{\max,\min}$ occur at slightly different values of the applied field H for the two orientations, they occur at the same values of B_r, and the sum in the second term of Eq. (5) is the same at the two orientations. Values for b can be determined from either the maximum or minimum values of the frequency modulations at the two orientations, and

$$b = \frac{\eta_1 - \eta_2 Q_{\max}}{1 - Q_{\max}} = \frac{\eta_1 - \eta_2 Q_{\min}}{1 - Q_{\min}} , \quad (8)$$

where

$$Q_{\max,\min} = \frac{(f_\alpha/f_\alpha^*)_{1\,\max,\min} - 1}{(f_\alpha/f_\alpha^*)_{2\,\max,\min} - 1} .$$

The frequency values shown in Table I were used with Eq. (7) to calculate values for a_γ and b which are shown in Table I. Values for b determined from Eq. (8) are also shown in Table I, and these values are consistent with those determined from Eq. (7). The uncertainties were calculated by the standard method of error propagation. Since it is a modulation effect we are measuring, there are actually two solutions for each data set: one with

b larger than and one with b smaller than the larger of the two η values with correspondingly different a_γ. Since the first three samples had approximately the same dimensions and demagnetizing factors, both solutions for b were approximately the same for all three samples, and which solution was correct could not be determined. Since the second solution gave a $b \simeq 0.6$, sample 4 was cut to have one of its demagnetizing factors with approximately this value. Then if this were the correct solution magnetic interaction effects would all but disappear along this direction. They did not, and the solution given in Table I is unambiguously the correct one. The large uncertainties in a_γ and b associated with sample 1-1A were caused by the substantial signal-to-noise ratio in these early data. The next largest uncertainties, associated with sample 4, were caused by the small difference in the two demagnetizing factors, thus offering "a shorter lever arm." Sample 3 represents the best quality and largest quantity of data. All the determinations for b are well removed from 1 and consistent from sample to sample. A value of $b = 1.25 \pm 0.05$ would seem to be reasonable. A more rigorous determination of b by numerically fitting the data making no approximations and explicitly including all three oscillations to an arbitrary number of harmonics does not change significantly the value found for b.[34]

IV. DISCUSSION

The experimental data interpreted within the framework of our empirical approach clearly yields a value for b greater than 1. The effects of experimental artifacts, such as the skin depth of the modulation field, a difference in the γ amplitude along the two $\langle 110\rangle$ directions, departure from ellipsoidal sample shape, inhomogeneity in the applied magnetic field, and detection sensitivity, have been considered and determined to influence our results to a degree well below that required to account for this discrepancy. The oscillation in the Fermi energy caused by the passage of a γ Landau level through it has also been considered. The resultant redistribution of the electrons among the quantum levels will indeed induce an oscillation in the α extremal cross section of the Fermi surface and cause a modulation of the α frequency with the γ period. However, the effect in Pb is to produce a b with a difference from 1 that is at least two orders of magnitude smaller than observed. No other mechanism of this type seems to exist to explain the anomalous value of frequency modulation.

Shoenberg and Vuillemin[15] (SV) made measurements of the frequency modulation in Au similar to those made here. They found that the observed frequency modulation of the high-frequency belly oscillation along [111] corresponded to an amplitude of the low-frequency neck oscillation that was some 30% larger than that measured macroscopically by a calibrated pickup coil, and they proposed that the difference was due to a variation in the magnetization over the sample. Such an interpretation would seem to be the most reasonable for this experiment as well. A difference in the magnetization averaged over the electron's cyclotron orbit and that averaged over the volume of the sample and appropriate for the demagnetizing field could account for our b parameter being different from 1,[35] and it allows for the retention of the basic tenet of the SC[36] that the electron "sees" the magnetic induction on the scale of its own orbit. The alternative is to discard the SC together with its theoretical justification, which is too speculative to be pursued. Since induction inhomogeneity seems the most likely case, there must be some mechanism or mechanisms that will produce a variation in M over a range at least on the order of the cyclotron orbit, so that the two averages will differ, and SV proposed that phase smearing due to crystal imperfections could produce such an effect. Watts and Coleridge[37] and Shoenberg[38] have considered this further, and indeed it is clear that the effect of such phase smearing is not negligible.

We now consider in a very qualitative way the effects of crystal imperfections. Dislocations can cause a local distortion of the lattice and produce a variation in the frequency of the dHvA oscillations from one part of the sample to another. The various portions of the sample then are always somewhat out of phase with each other, and the magnetization averaged over the sample is appropriately reduced. However, the magnetization averaged over the cyclotron orbit where the variations are not as large will not be reduced by as large a factor. The sizes of the cyclotron orbits in question here are approximately 1 μm and it is not at all unreasonable to expect that the dislocations in our samples are separated by this amount. If we adopt this view we immediately run into the problems associated with an induction that varies over the sample, because the magnetization varies. To make the problem manageable we will adopt the concept of a local demagnetizing factor η_r.[39] Then the field seen by the γ electron in carrying out its cyclotron orbit and appropriate for the dHvA effect is

$$B_r = H + 4\pi(1-\eta_r)M_{\gamma r} - 4\pi(\eta - \eta_r)\overline{M}_\gamma , \qquad (9)$$

where $M_{\gamma r}$ is the magnetization averaged over the cyclotron orbit and $\overline{M}_\gamma$ is that averaged over the sample. These two amplitudes are related to our empirical formalism by the approximate relation

$$b \approx \eta_r + (1 - \eta_r)(M_{\gamma r}/\overline{M}_\gamma) , \tag{10}$$

where equality is established by appropriately averaging the quantity on the right. It might be noted to a first approximation, neglecting local demagnetization, that b is an average measure of the ratio of the local to sample-averaged magnetizations.

The observed α oscillation will be the sum of the contributions from the various parts of the sample. The phase of the α oscillation will vary over the sample, because the α dHvA frequency varies with the lattice distortion and because B_r varies with $M_{\gamma r}$ as well. Shoenberg[38] has shown that the result depends on the degree of correlation that exists between the α and γ phase smearing, as well as on the distribution of phases over the sample. If a Lorentzian distribution of the γ phase is assumed and it is uncorrelated with the α phase distribution as well, it follows that to a first approximation

$$b^{-1} \simeq e^{-|\delta\phi_{\gamma 0}|} ,$$

where $|\delta\phi_{\gamma 0}|$ is the width of the γ-phase-smearing distribution. If the phase smearing is due to a frequency variation such as that which might be caused by a local strain distribution, then

$$\delta\phi_\gamma = \frac{2\pi f_\gamma}{B_r} \frac{\delta f_\gamma}{f_\gamma} ,$$

where δf_γ is the strain-induced variation of the γ frequency. Using the value of $b = 1.25$ together with the elastic constants of Pb (Ref. 40) and the hydrostatic pressure dependence of the dHvA oscillations determined by Anderson, O'Sullivan, and Schirber,[41] it has been determined that a lattice distortion on the order of $\Delta a/a = 3 \times 10^{-5}$ would account for this order of magnitude of phase smearing. Edge dislocations separated by the order of a micron could produce this lattice distortion.[42]

If the dislocations are indeed separated on a scale larger than the cyclotron orbits, a regional variation in the scattering of the electrons could also occur, and such a variation would produce an effect similar to the phase smearing caused by the shifting of the dHvA frequencies discussed above. This would result from a greater reduction of the γ amplitude in the region near the dislocation, with the corresponding shift in the α phase over that in the dislocation-free region. In addition, owing to the strong dependence of the α amplitude on the scattering, the contribution of the dislocation-free regions will be more heavily weighted in the determination of the α frequency modulation. If one assumes that the variation in magnetization is due entirely to inhomogeneous scattering and neglects the phase variation in the γ oscillation as well, the local amplitude $M_{\gamma r}$ may be characterized by a local Dingle term,

$$M_{\gamma r} = M_{\gamma 0} \exp - (q m_\gamma^* / B_r) X_{\gamma r} ,$$

with $X_{\gamma r}$ varying from region to region. There would be a similar term in each region for the α oscillation as well as the phase variation associated with the regional variation of $M_{\gamma r}$. The frequency modulation due to this depends on an appropriate averaging of $M_{\gamma r}$, which can be expressed by a properly averaged Dingle temperature $\overline{X}_{\gamma r}$ which is smaller than the corresponding $\overline{X}_\gamma$ appropriate for the volume-averaged $M_{\gamma r}$, i.e., $\overline{M}_\gamma$. If local demagnetization is neglected with $M_{\gamma r}/\overline{M}_\gamma \simeq b = 1.25$, a value of $\overline{X}_\gamma - \overline{X}_{\gamma r} = 0.14$ K results which is not at all unreasonable.

Finally the possibility of a mosaic substructure cannot be discounted. Since the γ surface is known to be reasonably tubular the result of such a substructure is straightforward to estimate, and an angular distribution on the order of 1° could account for the observed results.

We are presently working on a more refined approach than has been presented here which employs some of Shoenberg's phase-smearing concepts in a modified form and which will be published later. While a preliminary analysis indicates that the rough estimates made above are of the correct order of magnitude, detailed quantitative agreement has not yet been obtained. However, it is clear that phase smearing will affect both the frequency and amplitude modulations. The degree and phase relationship of the phase-smearing-induced amplitude modulation depends strongly on the correlation between the smearing of the high- and low-frequency oscillations. For example, if the two phase smearings are fully spatially correlated this contribution to the amplitude modulation and that of the FM-AM effect are in phase for negative (or anti-) correlation (one frequency increases from its unstrained value in the same region where the other frequency decreases) but π out of phase for positive correlation (both frequencies increase or decrease in the same region). For a total lack of correlation the phase-smearing-induced amplitude modulation occurs at one half the γ period for this strength of magnetic interaction.[43] Since the most likely situation is one of partial correlation,[44] which has not as yet been worked out, a reasonable estimate of the phase-smearing contribution of the amplitude modulation cannot be made. However, since most of the amplitude modulation is accounted for by the detection-system artifacts, some limit is placed on the amount of phase smearing that can be present.

In conclusion it may be safely said that in the case of Pb the dHvA amplitudes are larger at the level of the cyclotron orbit than those averaged over the volume of the sample, and plausible sources for such an inhomogeneity have been discussed. However, one puzzling aspect remains. The degree of magnetic interaction in SV's experiment was substantially below that of the experiment described in this paper, and the discrepancy in the amplitudes was correspondingly more difficult to establish. However, the difference in their amplitudes does seem to be outside the experimental uncertainty, and their results interpreted within our formalism yield a very approximate value for b of 1.3. If the discrepancy is caused entirely by crystalline imperfections it does seem strange that all of our samples produced the same effect and that SV obtained about the same value for this effect in Au that we have obtained for Pb.

Note added in proof. We have examined Shoenberg's phase smearing in some detail. We find that by treating the α and γ phase smearing as independently adjustable these experimental results can be accounted for with values of η_r that are near one third.

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[17]A. V. Gold, Philos. Trans. R. Soc. Lond. A 251, 85 (1958). What appears to be the first observation of a magnetic-interaction-related effect was made in this paper with the strong amplitude modulation of the high-frequency α oscillation along [110]. The α frequency value quoted was measured in the present experiment.

[18]The solution used for both etching and cutting consisted of 10%H_2O_2, 40%CH_3COOH, and 50%H_2O.

[19]The imperfect ellipsoidal shape yields a local variation of the demagnetizing factor. In sample 2, for example, the averaged demagnetizing field over the sample does not differ substantially from the value calculated assuming the ideal ellipsoidal shape, and expressed as an error on η, the absolute value of $\Delta\eta$ is not greater than 0.01 for either direction. More significant could be the effect of the field inhomogeneity due to the irregular shape, which yields a magnetization "smearing" of a sort even though of a different type than that introduced by local stress and microstructure. In sample 2 along the small demagnetizing direction, for example, the magnetization signal can be considered to be coming from the superposition of three "pseudocrystals": a crystal of about 95% of the actual volume with the ideal demagnetizing factor, $\eta = 0.068$, a second crystal of 4% of the actual volume with $\eta = 0.2$, and a third crystal with a 1% volume and $\eta \simeq 0.4$. Along the large demagnetizing directions the volumes of the three pseudocrystals are 95% at 0.828, 4% at 0.75, and 1% at 0.6. It is easy to see that this type of smearing will affect negligibly the modulation of frequency.

[20]P. M. Everett, Phys. Rev. B 6, 3553 (1972); 6, 3559 (1972).

[21]The low modulation level greatly enhances the higher-frequency α oscillation relative to the γ oscillation. In the examination of the γ amplitudes described above the field modulation amplitude was approximately 32 G, and the γ oscillation clearly dominated the signal.

[22]The magnet was calibrated with nuclear magnetic resonance.

[23]P. M. Everett, Rev. Sci. Instrum. 43, 753 (1972).

[24]Since the significant quantity is not the absolute frequency but rather the ratio of the apparent to the unmodulated frequency, each frequency value determined from the field values of alternate zero cross-

ings was divided by the average frequency determined over the field interval containing nine oscillations and with the oscillation of interest at its center. This procedure removed detectable drifting in the determined frequency modulation caused by nonuniformities in the sweep rate. A truly negligible scatter was introduced into our results by this procedure since the γ period contains 8.8 rather than 9 α oscillations. This procedure also eliminated any error that might have been introduced by an error in the determination of the absolute value of the α frequency.

[25]R. D. Plummer and W. L. Gordon, Phys. Lett. 20, 612 (1966).

[26]If the detected signal were skin depth limited, the portion of the sample contributing to the α signal should be substantially different in the [110] and [1$\bar{1}$0] orientations. As mentioned in Ref. 30 no significant difference was observed for the two orientations indicating that penetration of the modulation field was not a problem.

[27]By looking in quadrature to the signal and increasing the gain of the system, phase rolls of less than a degree should easily be detectable.

[28]S. Hornfeldt, J. B. Ketterson, and L. R. Windmiller, J. Phys. E 6, 265 (1973).

[29]HKW assumed an infinite rod-shaped sample which does not correspond to our ellipsoid. However, we should get a rough estimate of the effect at least for the field along the small demagnetizing direction.

[30]If one supposes for simplicity that the field inhomogeneity over the volume of the sample can be approximated by a Lorentzian distribution of 1 G width, the resultant phase-averaged γ amplitude will be reduced by less than 2% while that of the α oscillation will be reduced by about 16%. When the sample was placed in the homogeneous portion of the field no noticeable change in the α amplitude was observed at the node of the γ beat when the sample was rotated from [110] being along the field to [1$\bar{1}$0] along the field, i.e., with the thin and broad dimensions of the sample perpendicular to the field. This implies that the field inhomogeneities are almost perfectly matched for the two orientations, which would be most unlikely in an inhomogeneous retion of the field. However, as the sample was removed from the homogeneous region a marked difference in the α amplitude for the two orientations was observed but not for the frequency modulation.

[31]The calculation of Plummer and Gordon, Ref. 24, which has been extended to our situation, was developed for an infinitely long rod-shaped sample. Since this does not correspond particularly well to our ellipsoidal samples, there is a fair uncertainty in the determination of the magnitude of this contribution. However, this was not a point of major concern, because this contribution was fairly small to begin with and the amplitude modulation does not directly affect the frequency modulation in any case. It should be pointed out that the agreement with the observed amplitude modulation was slightly better along the small demagnetizing direction where the sample looks more rodlike.

[32]The formulas developed by Phillips and Gold (Ref. 13) can be used with the LK theory to predict the total harmonic amplitudes in the presence of magnetic interaction. The result depends on the spin splitting of the Landau levels, but unless the fundamental of the oscillation is very near the spin-splitting zero, which is known not to be the case, the third-harmonic amplitude is in the range of $(1-5)\times 10^{-3}$ of the fundamental amplitude. The fourth harmonic is down by another order of magnitude. The neglect of this amount of harmonic content will not have any substantial effect on our results.

[33]Under these conditions a_α is about 0.02 and no significant error will be introduced by neglecting the last term in Eq. (5).

[34]This procedure will be described in a future publication.

[35]Originally [Bull. Am. Phys. Soc. 19, 250 (1974)] we presented our results within a different formalism using an effective demagnetizing factor. There we assumed a difference between the local induction and that macroscopically averaged and appropriate for the demagnetizing field, and wrote $B_r = H_a + 4\pi M_r - 4\pi\eta\bar{M} = H_a + 4\pi(1-\eta')M_r$, with $\eta' = \eta\bar{M}/M_r$ the effective demagnetizing factor. This approach was suggested to us by Professor John Kimball. We switched to the b formalism (where, as is discussed below, b to a very rough approximation may be considered to be the ratio of M_r to $\bar{M}$), because we felt that in the long run it might prove more flexible. However, SV had already considered the basic idea encompassing a difference in the microscopic and macroscopic magnetizations which we missed owing to a lack of careful reading of their paper.

[36]Shoenberg conjectures that the electron Landau quantization is related to the magnetic induction. If one supposes uniform magnetization through the sample, the result $b' \neq 1-\eta$ will be in contradiction with Shoenberg conjecture.

[37]B. R. Watts, Philos. Mag. 24, 1151 (1971); P. T. Coleridge and B. R. Watts, Philos. Mag. 24, 1163 (1971).

[38]D. Shoenberg, private communication and J. Low Temp. Phys. 25, 751 (1976).

[39]Professor Shoenberg (Ref. 24) simultaneously suggested the use of η_r to us. Most of the following discussion is very qualitative in nature and is intended to relate our results to the phase-smearing concepts that Shoenberg and his colleagues have been developing over the past decade. Since the discussion is qualitative local demagnetization will be ignored, and η_r is presented here only for completeness.

[40]C. Kittel, *Introduction to Solid State Physics*, 4th ed. (Wiley, New York, 1971).

[41]J. R. Anderson, W. J. O'Sullivan, and J. E. Schriber, Phys. Rev. 153, 721 (1967).

[42]It is doubtful if the lattice distortions due to dislocations would be very isotropic, and the hydrostatic pressure variation of the γ dHvA frequency probably underestimates the phase smearing. However, this estimate was made for order-of-magnitude purposes only.

[43]This amplitude modulation becomes very complex for a factors above that appropriate to this experiment and well behaved for smaller a factors.

[44]As discussed in Ref. 38, W. M. Bibby's measurements on spherical Au samples suggest that the phase smearings of neck and belly oscillations are partially correlated.