curve in Ref. 3 is 0.56 Ry for two electrons per atom. But we deliberately raised it to 0.58 Ry so that our Fermi surface would have a closer resemblance to that in Ref. 16. The piece centered around P corresponds to the tetratube. Notice that the arm along P to H is pinched off in our version. The piece centered around H corresponds to the superegg. The susceptibility curve shows two peaks at the diameters of these two surfaces. As in the case of hcp rare earths, we expect the peak at the smaller wave vector (tetratube) to dominate because of the effect of wave-vector dependence of the matrix elements.[2] Hence, our calculation lends support to the idea that the nesting between the opposite faces of the tetratube is responsible for the helical structure of Eu. The superegg nesting anomaly should best be observed in the phonon spectrum of Eu.

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Landau-Level Widths, Effective Masses, and Magnetic-Interaction Effects in Lead*

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The amplitudes of the de Haas-van Alphen effect in lead have been studied in detail using large impulsive magnetic fields and employing a narrow-band filtering system. Crystals of high purity were grown, cut, and mounted in a manner which was essentially strain-free, and the Dingle broadening temperatures T_D for many of these crystals were found to be less than 0.1°K, corresponding to Landau-level widths of less than 1% of the level spacing $\hbar\omega_c$ in a field of about 10^5 G. These findings strongly suggest that the large level widths ($T_D \approx 1$–2°K) frequently reported in the past for many pure metals arise from destruction of phase coherence by dislocations. Accurate effective mass values have been determined for the extremal orbits at symmetry directions, and these results are compared with the data obtained directly from cyclotron resonance. The temperature dependences of the amplitudes for the harmonic and combination tones provide yet further detailed evidence for the correctness of Shoenberg's proposal that the Lifshitz-Kosevich theory must be modified when the magnetic interaction between conduction electrons is important. The absolute amplitudes of the fundamental oscillations are compared with the theoretical predictions, and the harmonic content is also discussed.

I. INTRODUCTION

IN studies of the de Haas-van Alphen (dHvA) oscillations in metals,[1,2] the major emphasis has usually been on the orientation dependence of the frequencies F which are directly related to extremal cross-sectional areas $\mathcal{A}_0$ of the Fermi surface. On the other hand, it is well known that studies of the amplitudes and waveforms of the oscillations can yield additional information about the conduction electrons in metals, in particular the cyclotron effective masses m^*, the effective widths k_BT_D of the Landau levels, and magnetic-interaction effects, but careful amplitude studies have been made for only a few metals so far. This paper is concerned with a detailed analysis of the temperature and field dependences of the dHvA amplitudes in lead, as well as with a comparison of the absolute values of the amplitudes and the harmonic content with the predictions of the basic Lifshitz-Kosevich (LK) theory.[3] In order to make a careful comparison of this kind it is necessary to know the shape of the Fermi surface in some detail, and we have chosen lead for this study since the shape of its Fermi surface is now well understood.[4]

One of the main objectives of this research was to make a comparison of the m^* values from the dHvA

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experiments with those obtained directly from cyclotron resonance (CR). However, it was necessary to limit our investigation to symmetry directions because of the large amount of time required for each precise determination of m^*. In the past, experiments on the temperature dependence of the dHvA amplitudes in lead as well as in several other metals have usually enabled m^* to be determined to about 10% at best, whereas in this work we have been able to reduce the error to about 1% by taking suitable precautions in the measuring technique. We also present the results of amplitude measurements for several harmonics as well as for some combination terms, and the amplitude variations for these nonfundamental terms provide further verification of Shoenberg's proposal[5] that the standard LK theory must be modified when the magnetic interaction between conduction electrons is important.

In Table I we give a summary of the rather meager results of previous work on the effective widths of the Landau levels for a variety of metals, as determined from the field dependence of the amplitude. The large broadening temperatures, with values of T_D often in excess of 1°K for relatively pure metals, have not been clearly understood in the past; for free electrons a value of 1°K for T_D corresponds to a level width which is as large as $\frac{1}{10}$ of the level spacing $\hbar\omega_c$ in a field of 75 kG. If it is assumed that the observed level broadening is caused by ordinary collisions with impurities and phonons, then we can find T_D from the relation[6,7]

$$T_D=\hbar/(2\pi k_B\tau), \qquad (1.1)$$

where τ, an appropriately averaged relaxation time, can be estimated from resistivity measurements. The values of T_D estimated in this way turn out to be some 10 to 100 times smaller than those found experimentally from the dHvA effect which implies that most of the observed level widths in Table I must be due to some other mechanism. In this paper we shall show that the Landau levels are actually very sharp in highly perfect crystals of pure lead, and indeed we believe that, for pure metals in which magnetic breakdown does not occur, any experimental values of T_D appreciably greater than 0.1°K can probably be attributed to the presence of dislocations in the sample.

TABLE I. Landau-level broadening temperatures T_D for various "pure" metals.

Metal	Reference	T_D(°K) dHvA expt.	T_D(°K) From resistivity
Ag	a	1.9	1.1
Al	b	1.5±0.5	···
Au	a	1.5	2.7
Be	c	4.0±0.5	···
Bi	d	1.5	···
Bi	e	4.0	···
Bi	f	1.5–2.2	···
Bi	g	0.2 (holes)	0.005
		0.65 (electrons)	0.014
Graphite	h	0.7–1.5	···
Graphite	f	1.5	···
Cu	a	1.3–1.7	0.5
Ga	a	1.0	0.02
Ga	i	1.3	<0.1
Hg	f	3.3±0.5	···
Hg	j	0.3 at 4.2°K	···
		2.0 at 1.2°K	···
K	h	0.4	0.02
K	k	0.4	0.02
Mg	l	0.7–0.9	0.25
Ni	m	$\lesssim$0.5	···
Pb	n	1.2–2.0	0.1
Pt	o	0.3–0.5	···
Sb	f	3.3±0.5	···
Sn	f	1.0–1.4	0.02
Zn	f	1.5	···
Zn	d	4.9	···
Zn	e	4.0	···
Zn	p	1.5±0.5	···

[a] Reference 5; see, also, P. E. King-Smith, Phil. Mag. **12**, 1123 (1965).
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II. DISCUSSION OF SOME THEORETICAL ASPECTS

A. Landau-Level Widths

Since the rather large values of $T_D\gtrsim 1$°K for pure metals cannot be accounted for in terms of ordinary collisions, we shall review briefly some other mechanisms for level broadening which have been suggested by a number of authors, namely intraband lattice broadening, magnetic breakdown, and the effects of dislocations.

Lattice broadening arises from the presence of the periodic crystal potential, which has the effect of splitting each discrete Landau level into a set of closely spaced sublevels. Any theoretical study of this splitting involves the difficult problem of solving the Schrödinger equation for a Bloch electron in a magnetic field. Early investigations of this problem[8–13] have shown that this

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[13] M. Ya. Azbel', Zh. Eksperim. i Teor. Fiz. **46**, 929 (1964) [English transl.: Soviet Phys.—JETP **19**, 634 (1964)].

form of broadening is likely to be minute except when the **k**-space orbit associated with the Landau level under consideration lies very close to either an open orbit or a self-intersecting orbit.

Zak[14] has recently reexamined this problem by using symmetry-adapted solutions of the free-electron Hamiltonian in a magnetic field as zero-order wave functions and then treating the lattice potential as a perturbation. Zak was able to show that for a simple cubic lattice the perturbation energy is given to lowest order by

$$V_n(\mathbf{k})=2V_{01}\exp(-\tfrac{1}{2}x)L_n(x)[\cos(2\pi m_1)+\cos(2\pi m_2)], \quad (2.1)$$

where $x=2\pi^2\hbar c/a^2eH$, H is the magnetic-field strength, a is the lattice constant, V_{01} is the first Fourier component of the lattice potential, $L_n(x)$ is a Laguerre polynomial, and m_1, m_2 are dimensionless mapping parameters which depend on the position of the electron in its **k**-space orbit. It is this dependence of the perturbation energy on **k** which results in a broadening of the Landau levels when the perturbation is averaged over one complete orbit. Assuming large values of n and x (n is the quantum number of the Landau level under consideration), Zak employs an asymptotic expansion for the Laguerre polynomials,

$$L_n(x)\approx\pi^{-1/2}\exp(\tfrac{1}{2}x)(xn)^{-1/4}\cos[2(nx)^{1/2}-\tfrac{1}{4}\pi], \quad (2.2)$$

to estimate the magnitude of the lattice broadening. Taking $n\sim10^3$ as being typical for large portions of the Fermi surface in a field of $H\sim10^5$ G and $x\sim10^5$ corresponding to $a\sim1$ Å, Zak finds that the spread of the perturbation (2.1) is about $10^{-2}\ V_{01}$, a result which is rather disturbing since it leads to a broadening which would actually *exceed* the level separation if $V_{01}\gtrsim0.1$ eV. However, Zak's numerical estimate of the broadening would seem to be incorrect since it appears[15] that the approximation (2.2) is valid only if $n>\frac{1}{4}x$, and this requirement is not satisfied for the assumed values of n, H, and x; in fact, an explicit computer calculation of (2.1) indicates that the perturbation should be less than $10^{-100}V_{01}$ under these conditions. Indeed Zak's result (2.1) for the lattice perturbation is completely in line with the findings of other authors since the condition $n=\frac{1}{4}x$ turns out to be precisely the condition that the nearly-free-electron orbits just touch the Brillouin-zone boundary in **k** space, i.e., the first open orbits are formed (along the cube axes).

Davis and Liu[16] have also considered these intraband broadening effects due to the lattice potential from the effective Hamiltonian point of view (see also Roth[17]). These authors were able to derive an explicit result for the level broadening and they conclude the the lattice broadening should be negligibly small unless the relevant extremal orbit lies within just a few (<10) Landau levels from an open orbit. Situations of this kind are expected to occur only rarely in practice, and the intraband lattice effect is thus unlikely to be responsible for the large values of T_D in Table I; for lead, specifically, we may safely state that there are *no* extremal orbits at symmetry directions which lie close to any open orbits. On the other hand it is well known that *interband* transitions, i.e., magnetic breakdown, lead to a broadening of the energy levels of the system since the effect of breakdown is to switch or scatter the electrons out of their orbits. The anomalous field dependence of the dHvA amplitudes which results from magnetic breakdown has now been investigated in a number of metals in which the band gaps are small,[18] whereas in lead we know that the band gaps are so large[4] that breakdown cannot occur in even the highest conventional fields; this conclusion is readily verified with the aid of Chambers' expression[19] for the breakdown probability in terms of known geometrical features of the Fermi surface.

Pippard[20] and Chambers[21] have suggested that the presence of just a few dislocations threading the orbits can have a profound influence upon the amplitudes of the dHvA effect since the Onsager quantization condition will no longer be strictly valid in a crystal which is subject to a nonuniform strain; the allowed orbital areas will then be slightly different for electrons moving through different regions of the metal. The problem of quantizing the orbits against the background of the complicated dislocation networks in real metals is evidently an insurmountable one, but it is perhaps not unreasonable to suppose that the net effect may be described heuristically in terms of a Lorentzian broadening of the Landau levels. Thus in the analysis of the present data we shall assume that the effects of the phase incoherence introduced by dislocations are contained in the experimentally determined parameter T_D.

B. Magnetic-Interaction Effects

A few years ago Shoenberg conjectured[5] that the magnetizing field appearing in the final LK result[3] should really be the total magnetic induction **B** rather than simply the applied field **H**, and this proposal has been justified by Pippard[22] on thermodynamic grounds. **M** should thus be determined self-consistently from

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TABLE II. Fourier coefficients in Eq. (2.7), ranked according to the order $O(i)$ of the various contributions.[a]

Term	$O(1)$	$O(2)$	$O(3)$	$O(4)$
p_1	A_1		$+\frac{\kappa A_1 A_2}{2\sqrt{2}}-\frac{\kappa^2 A_1^3}{8}$	
q_1			$\pm\frac{\kappa A_1 A_2}{2\sqrt{2}}$	
p_2		$A_2-\frac{\kappa A_1^2}{2\sqrt{2}}$		$+\frac{\kappa A_1 A_3}{\sqrt{2}}+\frac{\kappa^3 A_1^4}{6\sqrt{2}}-\kappa^2 A_1^2 A_2$
q_2		$\pm\frac{\kappa A_1^2}{2\sqrt{2}}$		$\pm\frac{\kappa A_1 A_3}{\sqrt{2}}\mp\frac{\kappa^3 A_1^4}{6\sqrt{2}}$
p_3			$A_3-\frac{3\kappa A_1 A_2}{2\sqrt{2}}$	
q_3			$\pm\frac{3\kappa A_1 A_2}{2\sqrt{2}}\mp\frac{3\kappa^2 A_1^3}{8}$	
p_4				$A_4+\frac{\kappa^3 A_1^4}{3\sqrt{2}}-\frac{2\kappa A_1 A_3}{\sqrt{2}}-\frac{\kappa A_2^2}{\sqrt{2}}$
q_4				$\pm\frac{\kappa A_2^2}{\sqrt{2}}\pm\frac{\kappa^3 A_1^4}{3\sqrt{2}}\pm\frac{2\kappa A_1 A_3}{\sqrt{2}}\mp 2\kappa^2 A_1^2 A_2$

[a] The subscripts refer to the harmonic number.

the implicit relation[23]

$$M=\sum_{r=1}^{\infty} A_r \sin\left(\frac{2\pi r F}{H+4\pi M}\mp\tfrac{1}{4}\pi-r\phi\right), \qquad (2.3)$$

where

$$A_r=-\frac{\nu T F}{H^{1/2}r^{1/2}}\left|\frac{\partial^2 \mathcal{A}}{\partial k_H^2}\right|_0^{-1/2} \times\cos(\tfrac{1}{2}rg\pi\mu)\exp\left[\frac{-r\lambda\mu(T+T_D)}{H}\right] \qquad (2.4)$$

is an approximation to the amplitude of the rth harmonic in the ideal LK theory for noninteracting electrons[24]; for simplicity we have taken **M** and **H** to be parallel and the sample is assumed to be in the form of a long rod parallel to **H** so that demagnetizing effects need not be considered. The cosine term in A_r arises from the spin splitting of the energy levels; μ is the cyclotron mass m^* expressed in units of the free-electron mass m; ϕ is a phase factor which will not concern us in this work; λ and ν are constants given by

$$\lambda=2\pi^2 mck_B/e\hbar=146.9\ \text{kG/°K},$$
$$\nu=4k_B(e/\hbar c)^{3/2}/(2\pi)^{1/2}=1.304\times10^{-5}\ \text{G}^{1/2}/\text{°K};$$

and $|\partial^2\mathcal{A}/\partial k_H^2|_0$ is a "curvature factor" (CF) which depends on the geometry of the Fermi surface in the neighborhood of the extremal area $\mathcal{A}_0$. Since $|4\pi M|\ll H$, it is clear that replacing H by B will hardly affect the slowly varying amplitudes A_r as such; however, even a small change in the magnetizing field can markedly affect the values of the sine terms, and the net result is that the amplitudes A_r' found by self-consistent solution of (2.3) may in fact differ appreciably from the ideal LK amplitudes A_r.

For $|4\pi M/H|\ll1$ we can write (2.3) as

$$M=\sum_{r=1}^{\infty} A_r \sin[r(x-\kappa M)\mp\tfrac{1}{4}\pi], \qquad (2.5)$$

where

$$x=(2\pi F/H)-\phi \quad\text{and}\quad \kappa=8\pi^2 F/H^2,$$

and (2.5) can be solved approximately to give M explicitly as a function of x; we consider here only the solution under conditions of a weak interaction with $|\kappa A_1|<1$, etc., which is indeed the case for the present measurements. Shoenberg[5] originally worked out the

[23] Throughout this paper M refers to the oscillatory component of the magnetization.

[24] For most of our experiments $\lambda\mu T/H\gtrsim2.5$ and the factor $1/\sinh(r\lambda\mu T/H)$ in the LK theory may then be replaced by $2\exp(-r\lambda\mu T/H)$ with negligible error. For the few cases where this approximation is not valid (in particular for the γ oscillations), appropriate corrections have been made throughout.

harmonic content (and modification of the fundamental amplitude) which is introduced when only the first term M_1 is considered on both sides of (2.5), and Girvan[25] has extended these results somewhat by using a Bessel-function expansion for the first three harmonic terms while still keeping only M_1 in the argument of the sine. However, if the cosine term in A_r happens to be small for the fundamental term, we expect to find the higher harmonics playing a role in determining the interaction effects, and thus it seemed desirable to seek a more general solution of (2.5) in which an arbitrary number of harmonic terms could be retained throughout. A general solution of this sort, in which M is given as an explicit function of x to any desired accuracy, can be found by means of a scheme of successive approximations in which we consider the nth approximation to M to be given by

$$M^{(n)}=\sum_{r=1}^{n} A_r \sin[r(x-\kappa M^{(n-r)})\mp\tfrac{1}{4}\pi], \qquad (2.6)$$

where we set $M^{(0)}=0$. Any expansion of (2.6) will evidently contain terms which involve various powers and cross products of the amplitudes A_r, and since the relative magnitudes of the A_r are determined largely by the exponential damping factors $\exp[-r\lambda\mu(T+T_D)/H]$, we can classify the various terms in orders $O(i)$ of small quantities according to the index i of the resulting negative exponentials; thus terms involving A_4, $A_1^2A_2$ or A_1A_3 will all be of $O(4)$. With this classification, the iterative scheme (2.6) is particularly simple in that any approximation $M^{(n)}$ is *exact* to $O(n)$; the next approximation $M^{(n+1)}$ merely adds to $M^{(n)}$ correction terms whose order is at least $O(n+1)$, as can be verified by inductive reasoning.

Various powers and cross products of sines and cosines are found in the expansion of (2.6) and by means of elementary trigonometrical manipulations we can express the nth approximation to M in the form of a harmonic series:

$$M^{(n)}=\sum_{r=1}^{n} [p_r \sin(rx\mp\tfrac{1}{4}\pi)+q_r \cos(rx\mp\tfrac{1}{4}\pi)]. \qquad (2.7)$$

The first few Fourier coefficients p_r and q_r can be found quite quickly and are given in Table II; the amplitudes $A_r'=(p_r^2+q_r^2)^{1/2}$ for the first three harmonics are then given in terms of the LK amplitudes A_r by

$$A_1'=A_1+0(3)+\cdots,$$

$$A_2'=A_2\left\{1-\frac{\kappa A_1^2}{\sqrt{2}A_2}+\frac{1}{2}\left(\frac{\kappa A_1^2}{\sqrt{2}A_2}\right)^2\right\}^{1/2}+0(4)+\cdots,$$

$$A_3'=A_3\left\{1-\frac{3\kappa A_1A_2}{\sqrt{2}A_3}+\frac{1}{2}\left(\frac{3\kappa A_1A_2}{\sqrt{2}A_3}\right)^2\left[1-\frac{1}{2}\left(\frac{\kappa A_1^2}{\sqrt{2}A_2}\right)\right]+\left(\frac{3\kappa^2A_1^3}{8A_3}\right)^2\right\}^{1/2}+0(5)+\cdots. \qquad (2.8)$$

Thus for $|\kappa A_1|<1$, etc., the effect of the magnetic interaction on the fundamental amplitude is expected to be small, the correction factor being of second order (relative to A_1) in exponentially small quantities, whereas for the harmonics the lowest-order interaction terms are of the *same* exponential order as the relevant amplitudes in the LK theory. The relative importance of the various interaction terms will depend not only on the Fermi-surface parameters $\mathcal{A}$ and the CF but also on the spin factor $\cos(\frac{1}{2}\pi rg\mu)$, and since in most cases the effective g values are not known, the magnitudes of the various dimensionless parameters in (2.8) cannot be determined in advance. If we consider the limiting case in which the dominant terms are those which involve only the fundamental amplitude A_1, we then recover Shoenberg's "strong-fundamental" results

$$A_1'=A_1+O(3)+\cdots,$$
$$A_2'=\tfrac{1}{2}\kappa A_1^2+O(4)+\cdots,$$
$$A_3'=\tfrac{3}{8}\kappa^2A_1^3+O(5)+\cdots,$$
$$A_4'=\tfrac{1}{3}\kappa^3A_1^4+O(6)+\cdots. \qquad (2.9)$$

In this case the temperature dependence of the rth harmonic should be given by $T^r\exp(-r\lambda\mu T/H)$ rather than by $T\exp(-r\lambda\mu T/H)$ as in the LK theory.

In the above analysis we have considered the amplitudes of M, whereas the measurements to be presented in Sec. IV actually refer to dM/dt rather than to M itself. Since M in (2.7) is an explicit function of H, the net effect of d/dt is to multiply A_r' by $2\pi rF\dot{H}/H^2$, which is just the (angular) time frequency ω of the oscillations. As we shall discuss later, the experimental conditions are such that ω is held constant for *all* harmonics during any set of measurements so that the amplitudes $\dot{A}_r'$ of dM/dt are simply given by $\omega A_r'$.

Modification of the harmonic content is not the only noticeable effect of the magnetic interaction. Another consequence of the nonlinear character of the interaction is that the crystal itself behaves like a "mixer" which can generate combination tones from the various fundamental terms which arise from different extremal sections through the Fermi surface, and we would expect the frequencies of the simplest of these mixed terms to be sums and differences of the fundamental frequencies.

[25] R. F. Girvan, U. S. Atomic Energy Commission Report No. IS-T-103, 1966 (unpublished). The expansion of (2.5) in terms of Bessel functions has also been pointed out by D. Shoenberg and J. J. Vuillemin, in *Proceedings of the Tenth International Conference on Low-Temperature Physics, Moscow, 1966*, edited by M. P. Malkov (Proizvodstrenno-Izdatel'skii Kombinat, VINITI, Moscow, 1967), pp. 67–84.

Thus if we consider the mixing together of just the fundamental oscillations from two extremal sections, we would have

$$M=M_a+M_b=A_a\sin\left[\frac{2\pi F_a}{H+4\pi(M_a+M_b)}-\phi_a\mp\tfrac{1}{4}\pi\right]$$
$$+A_b\sin\left[\frac{2\pi F_b}{H+4\pi(M_a+M_b)}-\phi_b\mp\tfrac{1}{4}\pi\right], \quad (2.10)$$

where the subscripts a and b refer to the two sections. Since $|4\pi M/H|\ll 1$, (2.10) reduces to

$$M=A_a\sin(x_a-\kappa_a M)+A_b\sin(x_b-\kappa_b M), \quad (2.11)$$

where

$$x_{a,b}=(2\pi F_{a,b}/H)-\phi_{a,b}\mp\tfrac{1}{4}\pi$$

and

$$\kappa_{a,b}=8\pi^2 F_{a,b}/H^2.$$

Now we have seen that for one frequency alone the amplitude of the fundamental term remains essentially unaffected if the magnetic interaction is weak so that replacing $M(B)$ on the right side of (2.11) by the ideal LK magnetization $M(H)$ should be a reasonable approximation as far as calculating the lowest-order combination terms is concerned. We then have

$$M=A_a\sin[x_a-\kappa_a A_a\sin(x_a)-\kappa_a A_b\sin(x_b)]$$
$$+A_b\sin[x_b-\kappa_b A_a\sin(x_a)-\kappa_b A_b\sin(x_b)],$$

and if we now assume that all four products κA are sufficiently small so that we may neglect all terms beyond the first order in these small quantities, the above equation can be solved approximately. The amplitude of the two simplest combination terms with frequencies $F_a\pm F_b$ are given to lowest order by

$$A_{a\pm b}=\tfrac{1}{2}A_a A_b(\kappa_a\pm\kappa_b), \quad (2.12)$$

so that the temperature variation of the amplitudes of these combination tones should be given by

$$A_{a\pm b}\propto T^2\exp[-\lambda(\mu_a+\mu_b)T/H]. \quad (2.13)$$

III. EXPERIMENTAL PROCEDURE

A. Sample Preparation

Since it was believed that dislocations might be the main cause of the large level broadening observed experimentally in metals of high purity, one of the aims of the present investigation was to determine if this broadening could be appreciably reduced by taking suitable precautions with the preparation of the samples. With these thoughts in mind, a considerable effort was devoted to problems associated with the growing and handling of very pure lead crystals of high crystallographic perfection. In what follows we give only a brief account of our crystal growing methods; a more detailed discussion of these techniques, as well as of the experimental procedure in general, can be found elsewhere.[26]

Our starting material was zone-refined lead (6NT Grade) obtained from Cominco Products Inc. Early experiments with crystals grown on a horizontal "Kapitza" furnace were found to give unsatisfactory results on two accounts; back-reflection Laue photographs of these crystals exhibited appreciable mosaic substructure, and the dHvA measurements of the effective broadening temperatures T_D were not reproducible from sample to sample. It was then found that the crystal quality could be substantially improved if the samples were prepared by the Czochralski method of pulling from the melt; very low dislocation densities have been found in crystals of other materials grown by this method.[27–29] All the lead crystals used for the measurements which are reported in this paper were grown by the Czochralski method at a rate of about 1 cm/h and in a vacuum of 10^{-6} to 10^{-7} Torr.

The pulled crystals were in the form of wires 3–6 cm long and $\lesssim$0.4 mm in diam, and it was necessary to develop a technique by which these soft, thin wires could be removed from the puller and subsequently cut into 6-mm lengths with minimum damage. The removal and cutting problem was finally solved with the help of a simple acid cutting technique. A beaker containing carbon tetrachloride to a depth of about 8 mm was placed under the crystal wire while it was still hanging vertically in the Czochralski apparatus, and the beaker was then slowly raised until a 7-mm length of the crystal was immersed in the liquid. A drop of a fast etchant[30] was added to the CCl_4, and this droplet slowly floated towards the wire and finally surrounded it at the liquid-air interface. The thin layer of etchant would cut through the lead wire in about 20 min, and the cut portion of the crystal would then sink slowly to the bottom of the beaker. After cutting, the 6-mm-long sample was carefully removed from the beaker, washed, and then gently guided into a small quartz trough, which was prepared by grinding a piece of thin quartz capillary along its length. Using the trough rather than the original capillary as a support for the crystal not only facilitated the mounting but also ensured excellent access of liquid helium to the sample. The crystal was held in the quartz trough with a tiny spot of either General Electric adhesive varnish or Corning stopcock grease; of the two, the grease was found to be preferable since the drying adhesive would often twist the sample slightly against the walls of the trough, thereby introducing some strain.

[26] R. A. Phillips, U. S. Atomic Energy Commission Report No. IS-T-170, 1967 (unpublished); a few numerical errors appear in this reference.
[27] R. S. Wagner, J. Appl. Phys. **29**, 1679 (1958).
[28] W. C. Dash, J. Appl. Phys. **30**, 459 (1959).
[29] S. Howe and C. Elbaum, Phil. Mag. **6**, 1227 (1961).
[30] 250 cc glacial acetic acid, 187.5 cc distilled H_2O, and 62.5 cc 30% H_2O_2.

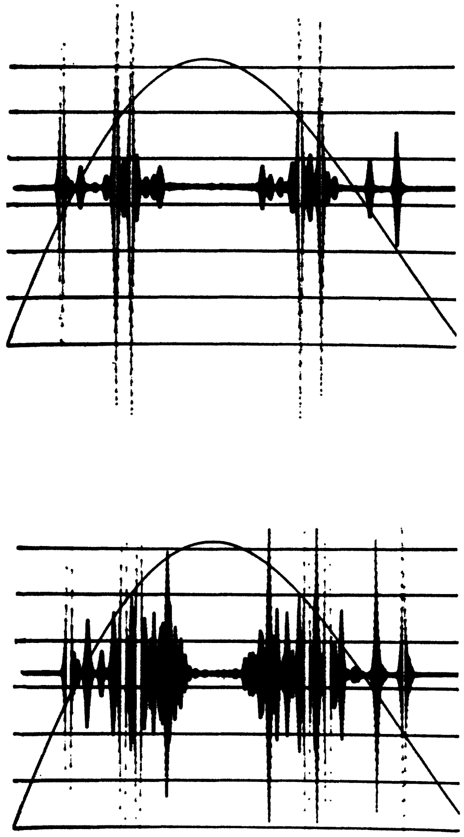

FIG. 1. Examples of the spectrometer action in impulsive fields for a [111] lead crystal. Peak field: 165 kG; pulse duration: 21 msec; filter: 135-kHz Butterworth 4-pole. Upper oscillogram: 3.5°K; lower oscillogram: 1.5°K.

The zero-field resistivity ratios $\rho_{300\,°K}/\rho_T$ for a typical crystal pulled by the Czochralski method were found to be 19 000 and 49 000 at 4.2 and 1.0°K, respectively. These values were determined from a series of careful resistance measurements in both longitudinal and transverse magnetic fields. From the absolute value of $\rho_{1\,°K}$ we estimate the mean resistivity relaxation time τ at 1°K to be 3.7×10^{-10} sec using the approximate relation

$$\tau\cong\left(\frac{12\pi^2\hbar}{ek_BS}\right)^2\frac{\gamma}{\rho_{1\,°K}},$$

where γ is the coefficient of the electronic specific heat and S is the total Fermi surface area,[4] and from (1.1) we find that this value of τ would correspond to a Dingle temperature $T_D\cong3$ m°K.

B. Impulsive Field Apparatus

The amplitude measurements were carried out between 1.0 and 4.2°K using the impulsive field technique developed by Shoenberg,[1,5] in which oscillations of the differential susceptibility are detected as an oscillatory emf induced in a pickup coil surrounding the sample. This induced voltage was displayed as one trace on a dual-beam oscilloscope and the field was measured by monitoring on the second beam the voltage developed across a standard resistor in series with the pulse coil; these two traces were simultaneously photographed on Polaroid film. Small portions of the discharge could be studied by expanding the time scale, and bucking techniques were used to obtain more accurate measurements of the field variations (see Ref. 4).

Partial separation of the many dHvA components was achieved with the aid of analog filtering as described in Ref. 26; for a given time frequency of the narrow-band detection circuit, each dHvA frequency F would appear as a resonance at different portions of the field pulse. The use of capacitors in parallel with the pickup coil was not sufficient for optimum analog resolution and two electronic band-pass filters, each attenuating at a rate greater than 12 dB per octave above and below the narrowing passing band, were used to improve the separation of the dHvA frequencies. The resolution was improved still further in later experiments by employing a pair of passive four-pole Butterworth filters operating in two ranges (center frequencies of 135 and 50 kHz and with 3- and 2-kHz bandwidths, respectively).[31] Some examples of the sharp frequency resolution achieved by the use of these passive filters are shown in Fig. 1. A stationary pickup coil was used throughout and the noise threshold of the entire detection system was determined to be less than 10 nV.

The pulse magnet was rigidly constructed and designed to produce a magnetic field which would be homogeneous to a few parts in 10^5 over the length of the sample. In order to reduce the radial inhomogeneity near the center of the coil, a coaxial conductor was used as the outside lead between the compensating windings; full details of the magnet construction are given in Ref. 26. The homogeneity of the magnet was checked with a pair of search coils as well as by direct NMR measurements, and the field was found to be homogeneous to 4 parts in 10^5 over a 9-mm length. Peak fields in excess of 200 kG were obtained when this magnet was used in conjunction with a 4320-μf capacitor bank which could be charged to 3 kV; the rise time was about 9 msec. The calibration of the magnet will be discussed in some detail in the following section.

C. Experimental Variables and their Control

Since the various dHvA terms are brought into resonance at the same time-frequency ω, it follows from (2.4) and the fact that $\dot{A}_1'=\omega A_1'\approx\omega A_1$, that the temperature and field dependence of any fundamental

[31] International Telephone and Telegraph Corporation, *Reference Data for Radio Engineers* (American Book—Stratford Press, Inc., New York, 1956), p. 191.

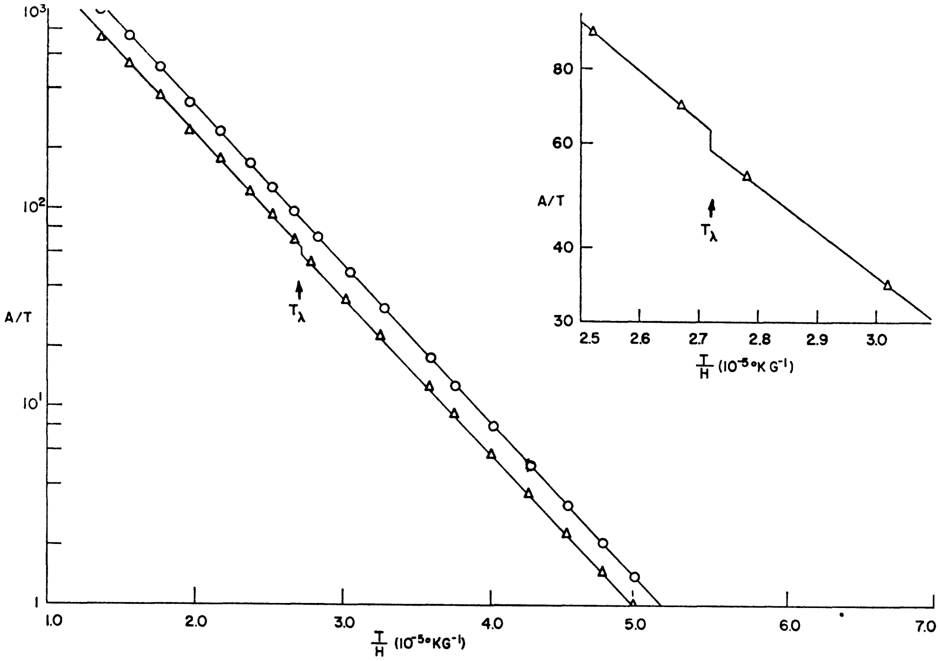

FIG. 2. Temperature dependence of the amplitude (arbitrary units) of the [100] β oscillations for $H=80.0$ kG, illustrating the discontinuity at T_λ due to hydrostatic-head effects. Δ, uncorrected results; $\circ$, corrected results.

amplitude in dM/dt should then appear only through the factor

$$TH^{-1/2}\exp[-\lambda\mu(T+T_D)/H]. \tag{3.1}$$

However, the experimental amplitude A is extremely sensitive to several other experimental factors, and these factors must be properly controlled or corrected for if the measured values of A are to be meaningful; here and in what follows we let A refer to all *experimentally measured* amplitudes in dM/dt. Most of the systematic errors which can arise in impulsive-field experiments have been discussed in some detail by Shoenberg,[5] and in this section we briefly summarize the precautions which we have taken to reduce them.

The temperature of the sample was determined from the vapor pressure of the He bath,[32] and since the helium level could not be monitored directly, the necessary hydrostatic-head corrections above T_λ were determined by means of a carefully calibrated germanium thermometer placed near the sample. The equilibrium sample temperature was found to exceed that indicated by the vapor pressure by as much as 40 m°K (just above T_λ). In Fig. 2 we illustrate the effects of this temperature error by plotting $\ln(A/T)$ against T/H for a typical series of fundamental amplitude measurements[33] between 1 and 4.2°K. A sharp discontinuity was found at T_λ when T was assumed to be given by the vapor pressure alone (lower curve; see also insert), whereas no discontinuity was found when the corrected values of T were used above T_λ (upper curve). The bath temperature was always lowered from one set of measurements to the next, and an adequate time interval (5–10 min) was allowed to ensure thermal equilibrium after each change in the vapor pressure. The temperature could be held constant to within 5 m°K for extended periods of time by using a simple manostat[34] to control the vapor pressure over the He bath. It is felt that with these precautions and the above corrections, the error in T should be less than 10 m°K.

The experimental amplitude A was taken to be proportional to the blip size at resonance and was measured directly from the Polaroid photographs with an average accuracy of 1%. Since A would often change by several orders of magnitude during a series of measurements, an accurate *relative* calibration of the various voltage ranges on the oscilloscope was made each time the range was changed, using an ac standard set to the particular resonant frequency being used. The actual field change appearing on the film represented only a small, but highly amplified, portion of the total field signal; most of the signal had been bucked out by an accurately known dc voltage. Calibration lines at intervals representing about 1% of the total field were displayed on the oscilloscope and superimposed on the photographs, and by interpolating between these lines the field value corresponding to the resonant blip could be read to an accuracy of about 0.2%. However, it was necessary to reduce the apparent readings because of overload recovery problems in the

[32] F. G. Brickwedde, H. van Dijk, M. Durieux, J. R. Clement, and J. K. Logan, J. Res. Natl. Bur. Std. (U. S.) 64A, 1 (1960).

[33] The abscissa in the temperature-dependence plots is chosen to be T/H rather than simply T since the slopes of the straight lines are then directly proportional to μ and independent of the (constant) field strength.

[34] The manostat was designed by Dr. C. A. Swenson.

oscilloscope amplifiers; in the bucking method, the amplifiers were driven beyond their operating ranges during most of the discharge. This correction was determined to be about 1% by recording the field pulse with a high-speed digital recording system[35,36] and superimposing time markers at each recording (every 400 μsec) on the field trace as it was being photographed. The corrected voltage readings were translated into field strengths using a conversion factor determined by dynamic calibration of the entire apparatus.[35] In the process the field readings were corrected by introducing an experimentally determined time delay $\tau_0 \approx 65$ μsec, a procedure which is described in detail by Panousis[36]; this correction is given by $\Delta H/H \doteqdot \pm\omega\tau_0 H/2\pi F$ (+ sign for falling field), and it was found in all cases that $\Delta H/H \lesssim 1\%$.

Further corrections must be applied to the apparent field values because of the "gliding-tone" effect, whereby the maximum response of a narrow-band detection system to a time-dependent driving frequency occurs at a value of the instantaneous driving frequency which differs from the natural resonant frequency of the circuit; this frequency difference corresponds to a shift δt in the time at which the maximum response occurs. According to Barber and Ursell[37] this time shift is proportional to a quantity $(a\omega)^{-1/2}$, and if we make the simplifying assumption that the amplitude of the driving dHvA oscillations in dM/dt is a constant, the results of Barber and Ursell can be translated into the language of our problem to give

$$\delta t = Cx^{-1/2}(\Omega^2 + 2\omega^2/x^2)^{-1/2},$$

where $x = 2\pi F/H$, ω and Ω are, respectively, the (angular) time frequencies of the natural resonance and the impulsive field (assumed to be an undamped half sine wave), and C is a constant of proportionality which depends only on the damping of the system. The time shift in turn corresponds to a systematic error δH in the field value assigned to the maximum amplitude; the fractional error in the field is approximately $\delta H/H = (\omega/x)\delta t$, the true field being greater than the apparent field under falling-field conditions.

In our experiments the resonant pickup circuit was followed by both active and passive filters of much narrower bandwidth than the width of the natural resonance of the pickup circuit alone. Since a detailed analysis of the response to time-dependent frequencies would be quite difficult, we have simply assumed that we can make use of the above functional dependence on x and ω (i.e., for a simple LCR circuit) as far as calculating the rather small field corrections is concerned. However, we have used the dHvA effect itself to make a dynamic determination of the constant of proportionality C. At [110] the γ oscillations exhibit a two-frequency beating pattern, and it was noticed that the field strengths corresponding to beat maxima and minima under conditions of narrow-band detection were different from those obtained using wide-band circuitry. From a study of these field shifts C was found to range from 1.35 to 1.95, depending on the particular experimental conditions used. $\delta H/H$ for the other dHvA frequencies was then found to range from 0.5% to 3.0%, the correction being least for the highest dHvA frequency F. Once the above corrections were made, it was felt that the net reliability of the field value assigned to the peak of any resonant blip was better than 1%.

Measurements of the temperature dependence of the amplitudes can be affected by small variations in the field at resonance which can occur from pulse to pulse due to heating in the magnet and to variations in the capacitor-bank voltage immediately before the discharge. Measurements of the resistance of the magnet immediately after a pulse showed that the magnet reached thermal equilibrium with the liquid-nitrogen bath in 2–4 min. With a 5–10-min waiting period between pulses and use of a highly stable power supply for charging the capacitors, variations in the field at resonance during any particular series of temperature-dependence measurements were found to be less than $\frac{1}{2}$%.

The dHvA amplitudes may be reduced if there is appreciable heating of the sample during the pulse. Shoenberg[5] has discussed the three main sources of heat production which might cause the temperature of the specimen to rise above that of the bath, namely, (a) eddy-current heating in the windings of the pickup coil; (b) reversible magnetocaloric effect in the specimen holder; and (c) eddy-current heating in the specimen itself. The possibility of heat transfer to the sample due to (a) is important only above T_λ and depends on the thickness of the liquid He layer between the sample and the former of the pickup coil. Rough estimates[5] indicate that the large bore (1.4 mm) of the pickup coil used in these experiments was more than adequate to prevent appreciable heat transfer to the sample from the pickup coil. The heating effects due to (b) were reduced by using quartz for the specimen trough, since according to Salinger and Wheatley[38] the magnetic susceptibility of quartz is a least 5 times smaller than that of Pyrex glass. By using split capillaries and thereby halving the volume of material, we estimate that the net heating of the sample due to the magnetocaloric effect in the quartz is at least an order of magnitude smaller than Shoenberg's estimates (<20 m°K below T_λ, <200 m°K above T_λ) for his Pyrex capillaries.

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[35] R. F. Girvan, A. V. Gold, and R. A. Phillips, J. Phys. Chem. Solids 29, 1485 (1968).

[36] P. T. Panousis, U. S. Atomic Energy Commission Report No. IS-T-175, 1967 (unpublished); P. T. Panousis and A. V. Gold, Rev. Sci. Instr. (in press).

[37] N. F Barber and F. Ursell, Phil. Mag. 39, 345 (1948). Shoenberg (Ref. 5) has given an explicit calculation of the gliding-tone effect for zero damping and for a resonance near peak field.

[38] G. L. Salinger and J. C. Wheatley, Rev. Sci. Instr. 32, 872 (1961).

Since the magnetoresistance in pure lead is fairly large, eddy-current effects in the thin samples used in these experiments are not expected to be serious. An explicit calculation of this effect is difficult but it is fair to say that the best indication of heating due to this or any other source would be the appearance of a *discontinuity* at T_λ in the temperature-dependence plots. However, no discontinuities were observed for most of the pure lead data although a slight amount of heating ($\sim$50 m°K) was noticed above T_λ in some of the [111] samples when very high peak fields were used [see the plots for the α_1 and δ_1 oscillations in Fig. 5(a)]. Since changes in the sample temperature of about 40 m°K could be easily detected when making the hydrostatic-head corrections, it is concluded from the smooth temperature-dependence plots that any temperature rise of the pure samples due to heating from any source was usually less than about 20 m°K. Further evidence for lack of heating effects comes from the fact that no significant difference was found in the results obtained from rising and falling fields. On the other hand, sizable heating effects ($\sim$200–500 m°K above T_λ) were observed in a Pb-0.64% Sn alloy, and in one particular run in which a grease block above the sample prevented good circulation of the liquid He above T_λ, the amplitudes were observed to increase by a factor of roughly 20 when the temperature was lowered below T_λ.

Accidental bending of the sample, due to mishandling or thermal cycling, can have a profound effect on the amplitudes and thereby lead to erroneous and irreproducible estimates of the broadening temperature T_D. Despite the precautions taken while growing, cutting, and mounting the samples, it could not be claimed *a priori* that any given specimen had not been slightly damaged, and thus a large number of samples ($\sim$60) were studied. As a further precaution, most of the measurements were made with field directions corresponding to turning points of the frequencies, where the effects of bending are expected to be least.[5] Inhomogeneities in the magnetic field over the dimensions of the sample can also modify the field dependence of the amplitudes appreciably, but such effects are not expected to be serious here since the inhomogeneity of the magnet was only a few parts in 10^5 over a 9-mm length. Care was always taken to center the sample in this region by moving the sample up and down in the cryostat and measuring the amplitude variations of several high-frequency oscillations. Under the conditions of the present experiments ($H > 50$ kG), it was found that the 6-mm long samples could be moved over a distance of about ± 3 mm from the "central" position without appreciable change in the amplitudes. Field inhomogeneity caused by eddy currents in the samples can likewise affect the amplitudes and the harmonic content in these impulsive-field experiments. According to Pippard[22] the ratio $\dot{A}_2/\dot{A}_1$ becomes modified by a factor which depends on the conductivity and on the product $r\sqrt{\omega}$, where r is the sample radius. By studying this ratio as a function of ω and r (by progressively thinning two thick samples) we have found that the observed ratio begins to increase when $r\sqrt{\omega} \gtrsim 2.5$ mm kHz$^{1/2}$, whereas $r\sqrt{\omega}$ was always less than this number for the experimental results reported in this paper. Further confirmation that eddy-current effects were usually negligible comes from the fact that the field dependences of the *fundamental* amplitudes were always found to be independent of ω even for the thickest samples used in this work. We should point out that any effects due to sample bending and to magnet inhomogeneity become reduced as the field strength is increased, and it is for this reason that the impulsive-field method is particularly suitable for studying the amplitudes of the dHvA effect.

When using the resonance method for studies of the field dependence of the amplitudes, one must consider possible field dependences which are associated with the static and dynamic response of the pickup and detection circuity. In the first place, the value of the resistance of the copper windings of the pickup coil was found to increase by 50% in a steady axial field of 60 kG so that one would expect the Q factor of the pickup system to be field-dependent. Secondly, the maximum response becomes reduced under gliding-tone conditions[5,37]; but it would be difficult to calculate the reduction reliably for the circuitry which we have used. We have once again adopted a more pragmatic approach and have carried out subsidiary experiments which were designed to reveal any spurious field dependences introduced by the detection circuitry itself.

In experiments performed in collaboration with R. A. Johnson, the sample was replaced by a tiny driving coil of the same geometry as a typical sample. This driving coil was energized by a current of constant amplitude, which was periodic in $1/H$ with adjustable frequency F so that the magnetic moment of the coil simulated a *constant-amplitude* dHvA effect; the energizing current was derived from a oscillator whose time frequency was controlled by a voltage derived from the magnet current. For constant detection frequency ω, it was found that the dynamic response of the entire detection and filter circuitry was essentially *independent* of changes in the field at which resonance occurred; this field could be changed by varying the pulse height, as was done in the field-dependence measurements of the actual dHvA amplitudes. Our experiments with the simulated signal thus indicate that no net field-dependent corrections need to be applied to (3.1) when analyzing the data. This finding was corroborated quite independently in actual dHvA experiments in which no changes in the field dependence were noticed when the effective Q of the pickup coil was reduced from about 20 to unity by means of an external shunt resistor, or when a different detection frequency ω was used for each set of field-dependence measurements. We might also mention that the resonance envelopes after the narrow-band filtering

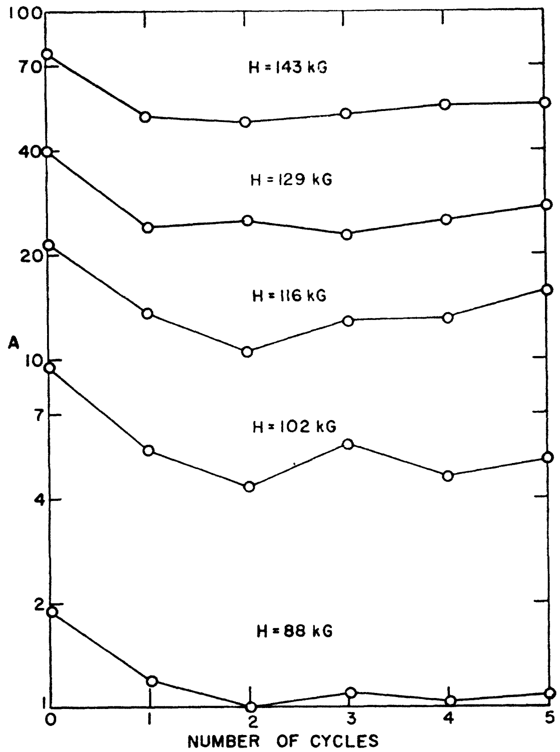

FIG. 3. Variation of the amplitude (arbitrary units) of the β_2 oscillations as a function of the number of times the crystal has been cycled between liquid-helium temperatures and approximately 150°K. The initial reduction and subsequent fluctuations were not observed in all crystals.

were nearly symmetrical and showed little evidence of the characteristic gliding-tone beat patterns.[5,37]

Although the amplitudes were completely reproducible throughout any run of several hours duration, they were not always reproducible from one run to the next. In fact, there were several instances in which the amplitudes were observed to decrease and then fluctuate after repeated thermal cycling between 1 and roughly 150°K, and an example of this cycling effect is shown in Fig. 3. In this example the amplitudes suffer a noticeable reduction at all field strengths after the first thermal cycle and we believe that this reduction is due to permanent damage of the sample incurred during the initial cooling as a result of surface strains introduced by the twisting of the sample against the walls of the trough. As long as the sample is maintained at liquid He temperatures, these surface strains would seem to be effectively prevented from moving into the bulk of the material and thus they should hardly affect the dHvA amplitudes. However, once the sample temperature has been raised, it seems possible that the surface strains migrate into the crystal and give rise to a loss in amplitude upon subsequent cooling. We cannot attribute the initial reduction in Fig. 3 to temperature changes *per se* since for several other samples the ampli-

TABLE III. Summary of effective masses in lead.

Orientation, oscillation[a]	F (MG)[b]	μ This work ($H\approx100$ kG)	μ Cyclotron resonance[c] ($H\approx5$ kG)
[100] α	208	1.51±0.03	1.58
[111] α	156	1.12±0.01	1.15
[110] α	159	1.10±0.01	1.12
[100] γ	~24.1	0.74±0.02	0.75
[111] γ	22.4	0.68±0.02	0.70
[110] γ	18.1	0.56±0.01	0.56
[100] π	36.0	0.89±0.02	0.92[d]
[100] β	51.4	1.22±0.01	1.23
[111] δ	110	1.19±0.01	1.20
[100] ϵ	~232	...	2.59
[111] ϕ	~365	3.2 ±0.1	...
[110] ω	...	...	1.26 or 1.41

[a] The assignments of the various oscillations to extremal orbits on the Fermi surface are given in Ref. 4.
[b] Most of the frequency values reported here are recent determinations made with an NMR probe in the immediate vicinity of the sample [J. R. Anderson, W. J. O'Sullivan, and J. E. Schirber (private communication)], and these results differ somewhat from those reported in Ref. 4. The frequency values preceded by a tilde (~) are from Ref. 4 but reduced by 5%.
[c] References 40 and 41.
[d] Assignment uncertain.

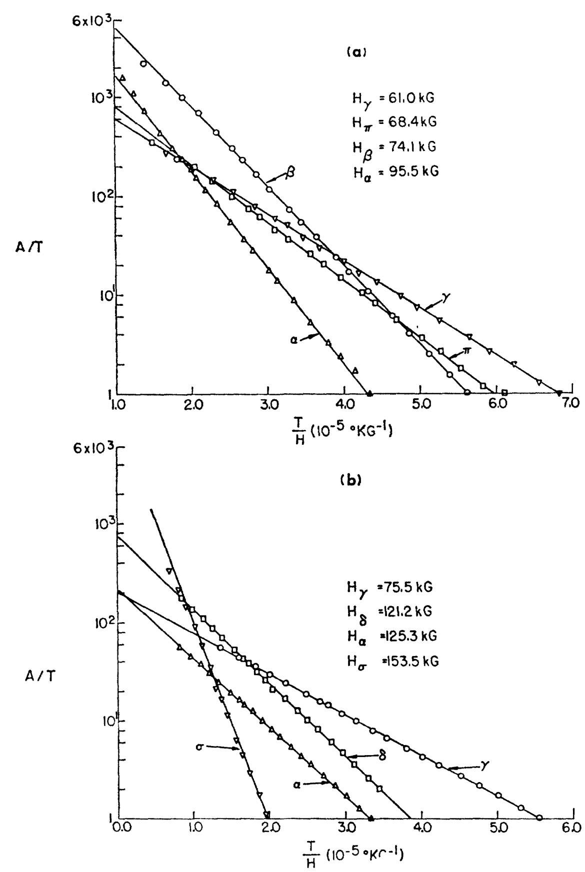

FIG. 4. Examples of the temperature dependence of the amplitudes (arbitrary units) of the fundamental oscillations for constant field H directed along (a), [100] and (b), [111]. The slopes of the lines are directly proportional to the effective masses μ.

tudes were found to be quite reproducible after thermal cycling. Most of the results in this paper were obtained from samples which were kept continuously at liquid He temperatures for several days while the entire set of measurements was being made, and in all cases the amplitudes were then found to be completely reproducible from day to day.

IV. EXPERIMENTAL RESULTS

A. Temperature Dependence

We first consider the temperature dependence of the fundamental amplitudes of dM/dt. If we assume that the results (2.8) are valid for lead, then the temperature variation of A_1' should be given by (3.1) so that for constant ω and H, a plot of $\ln(A/T)$ against (T/H) should yield a straight line[33] of slope $-\lambda\mu$. Typical data for the temperature dependence of the fundamental amplitudes[39] are presented in Fig. 4, and the excellent straight lines which were found for all the oscillations in lead indicate that the third-order correction terms in A_1' (see Table II) are of little consequence, at least within the range of fields used in these experiments (50–160 kG). The effective masses μ were found from least-squares determinations of the slopes, and the error in any one measurement of μ was estimated to be at most $1\frac{1}{2}\%$ as determined from the standard deviation ($<1\%$) of the slope and the uncertainty in the assignment of the field values at resonance. Weighted averages of the results from several samples are presented in Table III for $H\sim100$ kG; also included in Table III are the corresponding effective masses determined directly by Khaikin and Mina[40,41] from cyclotron resonance (CR) at $H\sim5$ kG. In all cases the 100-kG dHvA masses are found to agree well with the corresponding 5-kG CR masses. Even for the γ oscillations of lowest mass, the cyclotron frequency at 100 kG is not yet high enough relative to the dominant phonon frequencies to reveal any evidence of the field-dependent reduction in μ discussed by Fowler and Prange.[42]

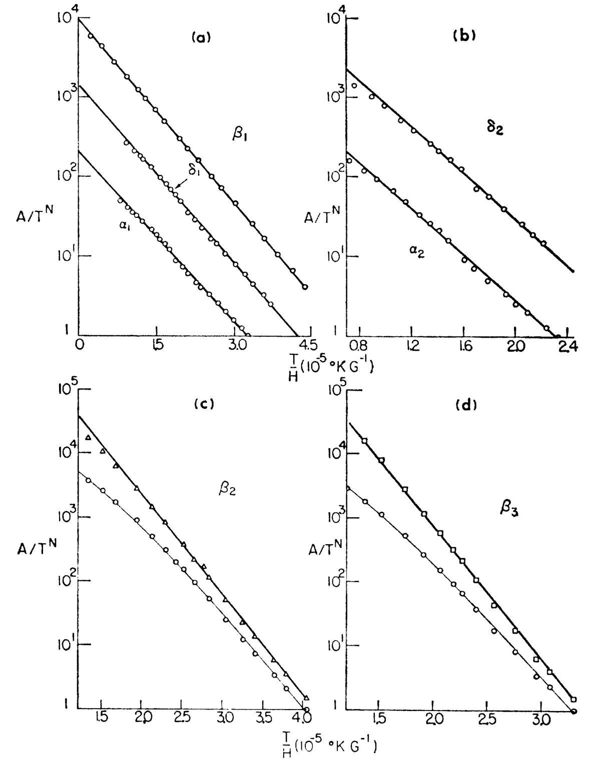

FIG. 5. Examples of the temperature variations of several fundamental and harmonic amplitudes (arbitrary units). The data points have been plotted according to the exponent N required by the LK theory ($N=1$) or by the strong-fundamental solutions (2.9) of the magnetic-interaction theory ($N=r$). ○, $N=1$; △, $N=2$; □, $N=3$. β oscillations, H∥[100]; α and δ oscillations, H∥[111]. The field strengths were as follows (in kG): β_1, 71.6; β_2, 79.5; β_3, 88.2; α_1, 127.3; α_2, 150.0; δ_1, 112.8; δ_2, 141.2.

Besides the temperature-dependence studies of the fundamental oscillations, a large amount of information was also gathered about the temperature variations of the numerous harmonic signals. The exact form of the temperature dependence of the harmonic amplitudes will depend on whichever term or terms in the square brackets of (2.8) are the dominant ones. If we assume that only one term is dominant, the temperature variation of the rth harmonic in (2.8) will be given by $T^N\exp(-r\lambda\mu T/H)$, where the index N is determined by the nature of the products or powers of the various amplitude factors in that dominant term; for example,

TABLE IV. Magnetic-interaction effects in the harmonic masses.[a]

Orientation, oscillation	N	μ_r	$r\mu$	H (kG)	$\omega/2\pi$ (kHz)
[100] β_2	2	2.50	2.44	79.5	145
β_3	3	3.67	3.66	88.2	145
[110] γ_2	2[b]	1.04	1.12	95.2	50
γ_3	1	1.50	1.68	110.2	50
γ_4	2	1.96	2.24	119.0	50
[111] γ_2	2	1.27	1.36	55.1	135
γ_2	2	1.30	1.36	99.4	50
γ_3	3	2.01	2.04	64.5	135
γ_3	1	1.92	2.04	111.1	50
α_2	1	2.28	2.24	150.0	135
δ_2	1	2.32	2.38	141.2	135

[a] The subscripts refer to the harmonic number r, and the values of the fundamental masses μ are from Table III, Col. 3. The integer N is the exponent of T in the ordinate A/T^N of the temperature-dependence plots; N is chosen to give straight lines in these plots with $\mu_r\approx r\mu$, and magnetic interaction effects are negligibly small when $N=1$.
[b] See text.

[39] See Ref. 4 for the correspondence between the Greek symbols for the various dHvA oscillations and the extremal orbits on the Fermi surface.
[40] M. S. Khaikin and R. T. Mina, Zh. Eksperim. i Teor. Fiz. 42, 35 (1962) [English transl.: Soviet Phys.—JETP 15, 24 (1962)].
[41] R. T. Mina and M. S. Khaikin, Zh. Eksperim. i Teor. Fiz. 45, 1304 (1963) [English transl.: Soviet Phys.—JETP 18, 896 (1964)].
[42] M. Fowler and R. E. Prange, Physics 1, 315 (1965).

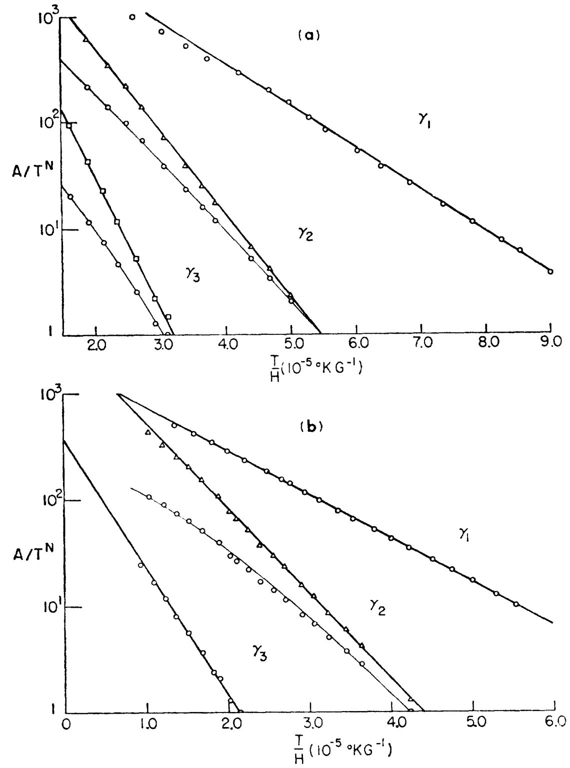

FIG. 6. Temperature-dependence plots for the amplitudes (arbitrary units) of the lowest harmonics of the γ oscillations for $\mathbf{H}\|[111]$. The data points have been plotted according to the exponent N required by the LK theory ($N=1$) or by the strong-fundamental solutions (2.9) of the magnetic-interaction theory ($N=r$). ○, $N=1$; △, $N=2$; □, $N=3$. The field strengths were as follows (in kG): (a) γ_1, 39.3; γ_2, 55.1; γ_3, 64.5; (b) γ_1, 76.2; γ_2, 99.4; γ_3, 111.1. Note the disappearance of magnetic-interaction effects for γ_3 when the field strength is doubled, i.e., N changes from 3 in (a) to 1 in (b).

we would have $N=2$ for terms involving A_1A_3 or A_2^2. Thus $N=1$ for the ideal LK theory (2.4), whereas $N=r$ for the limiting case of the strong-fundamental solutions (2.9), and intermediate values of N are also possible for the higher harmonics. It then follows that if N is chosen correctly, a plot of $\ln(A/T^N)$ against (T/H) will be a straight line with slope $-\lambda\mu_r$, where $\mu_r=r\mu$. Our experimental findings for the harmonic masses μ_r and the required values for the index N are summarized in Table IV, and we now proceed with a discussion of the results for the individual terms.

The temperature variations of the α_2 and δ_2 amplitudes for $\mathbf{H}$ parallel to [111] are presented in Fig. 5(b); for the sake of completeness we also show in Fig. 5(a) the fundamental amplitude variations for the α_1 and δ_1 oscillations.[43] Good straight lines are obtained for both harmonics by setting $N=1$, and the slopes of the lines yield values for the harmonic "masses" μ_2 which are very close to *twice* the respective fundamental masses μ, i.e., μ_2 and μ scale by the harmonic number $r=2$. The choice $N=1$ thus appears to be the correct one, which implies that the magnetic-interaction effects are of negligible importance for these particular oscillations, at least for the high field strengths used here.

The temperature dependence of the β_2 amplitude for $\mathbf{H}$ parallel to [100] is presented in Fig. 5(c). When the results for β_2 are plotted assuming $N=1$, the light curve drawn through the points exhibits considerable curvature. On the other hand, a plot of the data assuming $N=2$ results in a good straight line (heavy curve) and the slope of this line yields a value for μ_2 which is very nearly equal to $2\mu=r\mu$. Thus the choice $N=2$ appears to be the correct one, which implies that Shoenberg's strong-fundamental results (2.9) are appropriate. In Fig. 5(d) we show the temperature variation of the β_3 amplitude at the same orientation. As for β_2, the light curve for $N=1$ shows considerable curvature, whereas the heavy curve for $N=3$ is an excellent straight line, and the effective mass μ_3 calculated from the slope of this line is almost exactly $3\mu=r\mu$. Thus the limiting results (2.9) are found to be obeyed to high accuracy for the β_1, β_2, and β_3 amplitudes, and this

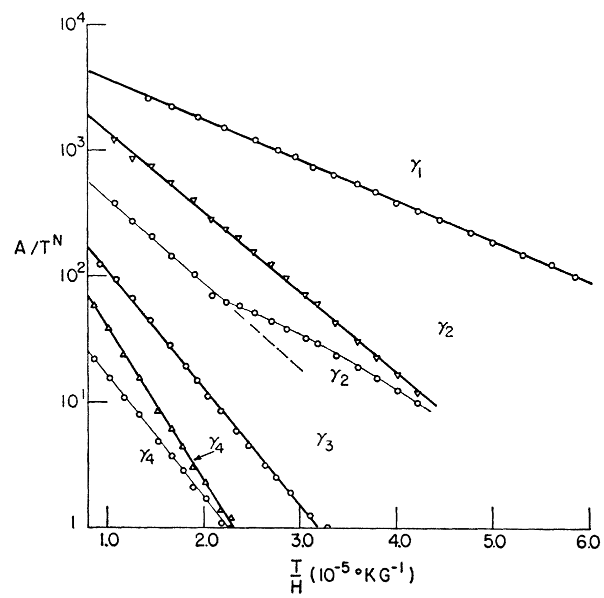

FIG. 7. Temperature dependence of the harmonic amplitudes (arbitrary units) for the γ oscillations for $\mathbf{H}\|[110]$. The data points have been plotted according to the exponent N required by the LK theory ($N=1$) or by the strong-fundamental solutions (2.9) of the magnetic-interaction theory ($N=r$). ○, $N=1$; △, $N=2$; ▽, $N=2$, as explained in the text. The nonlinear temperature dependence for γ_2 shown by the light curve for $N=1$ appears to reflect a transition from the interaction behavior at high temperatures to the ideal case at low temperatures, which can be accounted for by taking higher-order interaction terms into account. *Harmonic*-interaction effects are apparent in the data for γ_4 since the correct slope is obtained by choosing N to be 2 rather than 1 or 4. The field strengths are given in Table IV.

[43] Heating effects become noticeable above T_λ for $\mathbf{H}\|[111]$ and at the high values of the peak field (and hence of dH/dt) which were necessary for studying the harmonic amplitudes; no heating effects are apparent for the data in Fig. 4(b) which were obtained for much lower peak fields.

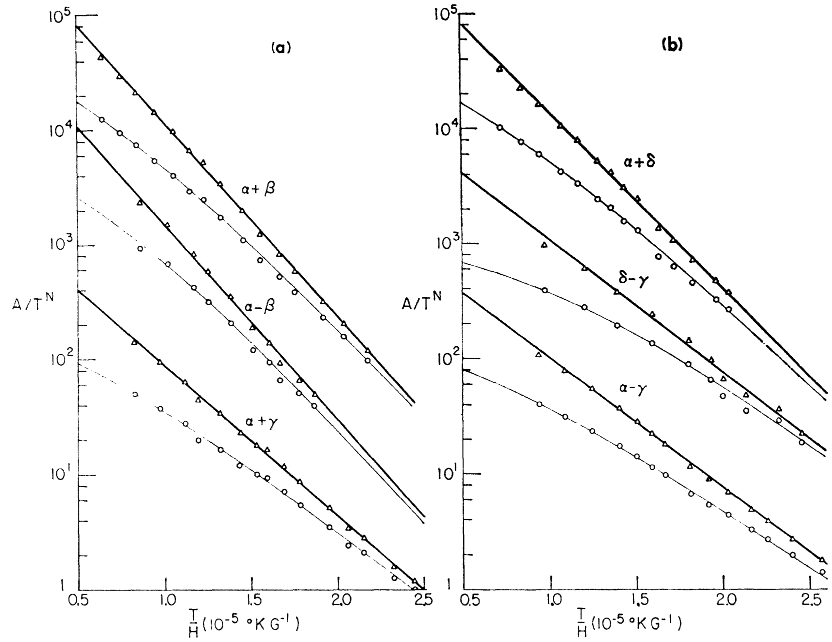

FIG. 8. Temperature dependence of the amplitudes (arbitrary units) for several combination oscillations for constant field H directed along (a) [100] and (b) [111]. ○, $N=1$; △ $N=2$, as required by the magnetic-interaction theory in its simplest form. The field strengths were as follows (in kG): (a) $\alpha-\beta$, 119.2; $\alpha-\gamma$, 125.3; $\alpha+\gamma$, 156.8; (b) $\delta-\gamma$, 104.6; $\alpha-\gamma$, 134.1; $\alpha+\delta$, 147.0.

conclusion is fully supported by measurements of the harmonic content (to be discussed later).

Thus far we have been able to account for the temperature dependences of the harmonic amplitudes in terms of the two extreme cases, the ideal LK theory (α and δ oscillations) and the strong-fundamental interaction theory (β oscillations). On the other hand, a variety of regimes appear in the temperature dependences of the data for the harmonic amplitudes of the γ oscillations. Because of the low values of μ for these oscillations, it was necessary to take into account the full sinh dependence of the amplitude coefficients[24] and the appropriate corrections have been applied throughout when making the A/T^N plots. These corrections are largest for the fundamental oscillations but can also lead to serious modifications for the harmonic amplitudes if interaction effects are important since various powers of A_1 appear in the interaction results.

The temperature dependences for the fundamental and harmonic amplitudes of the γ oscillations at [111] are presented in Fig. 6 for two different field strengths. In Fig. 6(a) the $N=1$ plot for γ_1 shows noticeable deviation from a straight line at low T, whereas at the much higher field strength used for the data shown in Fig. 6(b) there are no significant deviations from linearity, and it is believed that the apparent amplitude reduction in Fig. 6(a) is due to slight eddy-current damping at this low field strength. Turning now to the γ_2 oscillations at [111], excellent straight lines were obtained at both field strengths by setting $N=2$, and for both plots the experimental values for μ_2 are close to 2μ, indicating that the results (2.9) are appropriate. The two plots for the γ_3 oscillations are of special interest since two different values of N, depending on the field strength, are required if the slopes are to be similar with $\mu_3 \approx 3\mu$. Thus for low H we find the temperature dependence of the γ_3 amplitudes satisfying an $N=3$ plot while for large H the ideal theory with $N=1$ appears to be valid.

In Fig. 7 we show the temperature dependences of the various harmonic amplitudes of the γ oscillations at [110]. The data for the second harmonic γ_2 exhibit a marked change in slope in the plot for $N=1$ (light curve), the slope of the *low*-temperature data being consistent with $\mu_2 \approx 2\mu$. This suggests that the departure from linearity at the higher temperatures might be due to a change in the relative importance of the various interaction terms, and indeed we have been able to achieve a linear fit to the γ_2 data with $\mu_2 \approx 2\mu$ over the entire temperature range (heavy curve for $N=2$) by taking into account the A_1^4 terms in the fourth column of Table II in addition to all the second-order terms in the second column. Although this explanation in terms of higher-order interaction terms seems plausible enough, it requires a definite choice of values for the temperature-independent parts of the two parameters $\frac{1}{3}\kappa^2 A_1^2$ and $\kappa A_1^2/\sqrt{2}A_2$. It is impossible to comment on whether our choice was reasonable because of lack of information about the relevant g factor and because further higher-order terms should probably be included. The linear fit to the γ_3 data for $N=1$ in Fig. 7 with $\mu_3 \approx 3\mu$ confirms that the strong-fundamental approximation is not valid for the γ oscillations at [110], and indeed it was necessary to choose $N=2$ for the temperature dependence of the γ_4 amplitude in order to obtain $\mu_4 \approx 4\mu$. This latter result suggests that the *harmonic*-interaction terms are the important ones for

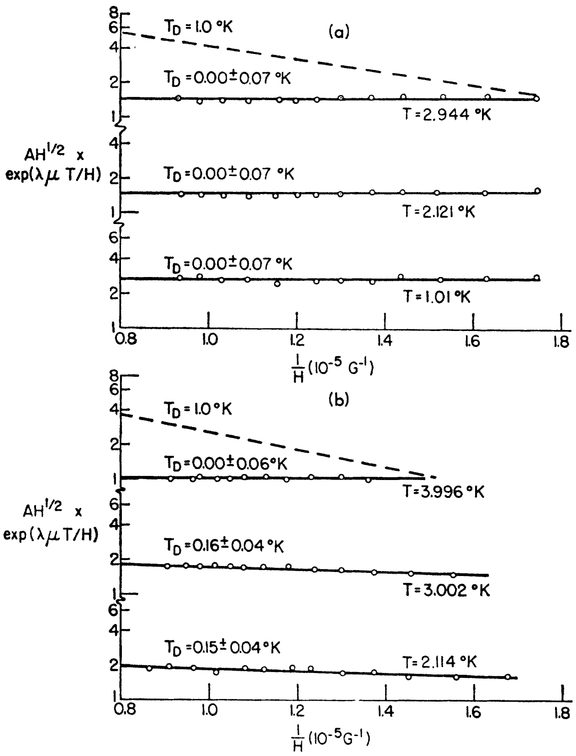

FIG. 9. Field dependence of the amplitudes of (a) the π oscillations at [100] and (b) the δ oscillations at [111] for several bath temperatures. The dashed curves indicate the slopes expected for a broadening temperature of 1°K.

the γ_4 oscillations at [110], with the $A_2{}^2$ term in Table II most likely being the dominant one.

Finally, we consider the temperature dependences of the amplitudes of the most prominent of the combination terms. According to (2.13) a plot of $\ln(A/T^N)$ against (T/H) should result in a straight line if $N=2$, and this is indeed borne out by the data in Fig. 8. The apparent effective masses $\mu_{a\pm b}$ found by dividing the slopes of these plots by λ are given in Table V, and as is predicted by (2.13), the values of $\mu_{a\pm b}$ are found to be in close agreement with the *sums* of the masses of the two fundamental components; in particular, we have been able to verify that $\mu_{a+b}=\mu_{a-b}$ for two pairs of sum and difference terms.

TABLE V. Apparent effective masses $\mu_{a\pm b}$ for the sum and difference combination terms.[a]

Orientation, oscillation	$\mu_{a\pm b}$	$\mu_a+\mu_b$	H (kG)	$\omega/2\pi$ (kHz)
[100] $\alpha-\gamma$	2.10	2.25	125.3	145
$\alpha-\beta$	2.72	2.73	119.2	145
$\alpha+\beta$	2.71	2.73	157.1	145
[110] $\alpha+\gamma$	1.61	1.66	134.4	50
$\alpha-\gamma$	1.61	1.66	135.1	50
[111] $\alpha-\gamma$	1.79	1.80	134.1	135
$\delta-\gamma$	1.85	1.87	104.6	135
$\alpha+\delta$	2.42	2.31	147.0	135

[a] μ_a and μ_b are the appropriate fundamental masses from Table III, Col. 3.

TABLE VI. Summary of maximum and minimum broadening temperatures for pure lead.[a]

Orientation	Oscillation	T (°K)	$\omega/2\pi$ (kHz)	T_D (°K)
[100]	Sample Pb-100-60A (smallest T_D)			
	π	1.01	50	-0.01 ± 0.07
	α	2.94	145	-0.09 ± 0.07
	β	1.04	81	0.08 ± 0.05
	Sample Pb-100-50B (largest T_D)			
	π	1.04	50	0.98 ± 0.06
	α	2.33	145	0.87 ± 0.14
	β	1.03	81	0.41 ± 0.05
[110]	Sample Pb-110-11 (smallest T_D)			
	α	3.99	135	-0.01 ± 0.07
	Sample Pb-110-10 (largest T_D)			
	α	4.18	145	0.26 ± 0.20
	γ	1.01	50	0.30 ± 0.10
[111]	Sample Pb-111-10 (smallest T_D)			
	α	4.00	135	-0.01 ± 0.09
	δ	4.00	135	-0.02 ± 0.06
	Sample Pb-111-1 (largest T_D)			
	α	4.16	100	$0.84+0.14$
	δ	4.00	100	0.68 ± 0.10

[a] The values of T_D quoted for the β and γ oscillations could be in error by a factor of 2 due to the complicated beat character of these oscillations. A complete listing of the individual results from over 20 samples is given in Ref. 26.

B. Field Dependence

We now turn our attention to the field dependence of the fundamental amplitudes. As we have seen, the *fundamental* amplitudes are not noticeably affected by interaction effects, and thus according to the arguments presented in Sec. III C the field variation of A_1' should be given by (3.1) without further modification, i.e., for constant ω and T, a plot of $\ln[AH^{1/2}\exp(\lambda\mu T/H)]$ against $1/H$ should be a straight line of slope $-\lambda\mu T_D$. Linear plots were indeed found for most of the crystals, excluding, of course, the data for the β and γ oscillations which exhibit an intrinsic beat structure (660 and 42.5 cycles per beat for β at [100] and for γ at [110], respectively), and representative field-dependence plots are shown in Fig. 9. The probable error for each determination of T_D was estimated from the standard deviation of the slope and the uncertainties in the absolute values of μ, T, and H, and the maximum and minimum values of T_D found for each set of oscillations are summarized in Table VI.

Although T_D was found to vary from sample to sample, the lowest values for each set of oscillations were found to be essentially zero within an accuracy of less than 0.1°K, whereas we have seen that the low-temperature broadening due to ordinary collisions is expected to amount to only 0.003°K and is thus eclipsed by our experimental uncertainty. Since the corresponding Landau-level widths for the better samples turn out to be less than 1% of the level separation $\hbar\omega_c$ for $H\sim100$ kG, we conclude that the large spread of T_D values found for samples pulled from the same melt arises

TABLE VII. Absolute amplitudes $|\kappa A_1|$ of the fundamental oscillations at 100 kG and 1.0°K.

Orientation, oscillation	$\|CF\|$[a]	$\omega/2\pi$ (kHz)	T_D (°K)	$\|\kappa A_1\|$[b]	$\|\cos(\frac{1}{2}\pi g\mu)\|$[c]	Possible g values[d] for $1.5<g<6.0$
			Third-zone electron surface			
[100] π	2.75	50	0.00	0.011	0.14	3.27, 3.48, *5.52*, 5.72
		50	0.00	0.012	0.15	3.26, 3.48, *5.50*, 5.73
β	0.07	50	0.08	0.33	0.33	2.28, 2.63, 3.92, 4.27, *5.56*
		50	0.15	0.33	0.30	2.30, 2.62, 3.94, 4.26, *5.58*
[111] δ	7.20	135	0.00	0.12	0.39	2.32, 2.71, 4.00, 4.40, *5.68*
[110] γ	2.30	50	...	$\lesssim$0.14[e]	...[f]	
[111] γ	...	50	$\sim$0.00	$\sim$0.34	...[f]	
			Second-zone hole surface			
[100] α	44.8	135	0.08	0.045	0.21	1.90, 2.08, 3.22, 3.40, 4.55, *4.72*
		145	0.08	0.033	0.16	1.92, 2.06, 3.24, 3.38, 4.57, *4.70*
		145	0.00	0.028	0.11	1.94, 2.03, 3.26, 3.36, 4.58, *4.68*
[110] α	22.0	135	0.00	0.11	0.28	2.56, 2.89, 4.38, *4.71*
[111] α	6.00	135	0.00	0.11	0.16	2.59, 2.86, 436, *4.65*

[a] CF values computed from a revised model for the Fermi surface which is based on a nonlocal pseudopotential [J. R. Anderson (private communication)]; these values differ from those obtained from the simpler model of Ref. 4.
[b] The values of $|\kappa A_1|$ were measured at different field strengths, and the field dependence of the LK expression (2.4) has been used to adjust them to a common field strength of 100 kG.
[c] Required to give exact agreement between theory [Eq. (2.4)] and experiment.
[d] The italicized g values are the lowest ones which appear to be common to all orbits on each sheet of the Fermi surface, while the lowest g values fo $g>1.5$ are those for which the product $g\mu$ takes on its lowest common value for *all* orbits (between 2.78 and 2.93).
[e] Value of $|\kappa A_1|$ at $H=94.6$ kG, which is the field at which the γ_2 amplitude was measured (Fig. 7).
[f] Detailed comparison with theory hampered by complex beat structure.

from the introduction of appreciable dislocation densities in some crystals but not in others, as discussed in Secs. III A and III C. Our findings are thus in keeping with the idea that broadening due to the lattice potential is negligibly small in metals for which magnetic breakdown does not occur. Unfortunately, we have not been able to make a direct correlation of the observed T_D values with the actual dislocation densities since the standard metallurgical techniques for counting dislocations do not seem to be immediately applicable to our thin lead samples. We should also point out that T_D was found to be independent of temperature between 1 and 4°K, as illustrated for the π oscillations in Fig. 9(a). However, as is shown in Figure 9(b) for the δ oscillations, a slight temperature dependence was found for the high-frequency oscillations at [111], but this apparent variation is believed to be due to either eddy-current effects[43] or the presence of strong combination terms whose frequencies are close enough to the fundamental frequencies to introduce a spurious temperature dependence in T_D for the fundamental term (e.g., the frequency of the terms $\alpha-2\gamma$ and $\alpha-\gamma_2$ differs by less than 1% from that of the δ oscillations).

C. Absolute Amplitudes and Harmonic Content

We now turn to the measurements of the absolute amplitudes κA_1 of the fundamental oscillations in $4\pi dM/dB$. The amplitude of the observed resonant blip can be expressed as the voltage

$$V=K\left|4\pi\frac{dM_1}{dB}\right|\frac{dB}{dt}\cong K\left|\kappa A_1\right|\frac{\omega H^2}{2\pi F},$$

where the conversion factor K takes into account the sample volume, effective number of turns of the pickup coil, end effects, and the effective response of the entire circuitry. It was possible to determine this factor dynamically with the aid of the simulated constant-amplitude signal described in Sec. III C since the driving coil was designed to have approximately the same dimensions as those of a typical sample.[44] The observed blip height from the test signal of frequency F, measured at the same time as frequency ω, then becomes $V'=K|4\pi dM'/dt|$, where $M'=nI/c$ is the equivalent magnetic moment per unit volume associated with a current I flowing in a coil with n turns per unit length. Thus the absolute amplitude is given by

TABLE VIII. Absolute amplitudes of combination oscillations.[a]

Orientation, oscillation	H (kG)	$\omega/2\pi$ (kHz)	T (°K)	$\|4\pi dM_{a\pm b}/dH\|$ Observed	$\frac{1}{2}A_aA_b\kappa_{a\pm b}$ From observed A_a, A_b
[100] $\alpha+\beta$	94.4	145	2.1	2.4×10^{-3}	2.2×10^{-3}
$\alpha-\beta$	90.4	145	2.1	1.0×10^{-3}	0.6×10^{-3}
[110] $\alpha+\gamma$	134.4	50	4.0	3.8×10^{-3}	2.2×10^{-3}
$\alpha-\gamma$	135.1	50	4.0	3.2×10^{-3}	1.4×10^{-3}
$\alpha+\gamma$	134.4	50	1.0	5.6×10^{-2}	3.4×10^{-2}
$\alpha-\gamma$	135.1	50	1.0	5.3×10^{-2}	2.1×10^{-2}
[111] $\alpha+\delta$	147.0	135	3.6	5.0×10^{-4}	3.6×10^{-4}
	147.0	135	1.0	1.4×10^{-2}	1.0×10^{-2}

[a] The field dependence of the LK expression (2.4) was used to adjust the observed values of the relevant fundamental amplitudes A_a and A_b to correspond to the field strength H at which the amplitude of the combination term was measured; in so doing, allowance has been made for the beats in the β and γ oscillations.

[44] The driving coil was 5.74 mm long and consisted of 179 turns of No. 50 A. W. G. wire wound on a mandrel of 0.34-mm diam.

TABLE IX. Harmonic content for α, β, and δ oscillations.

Oscillation	T (°K)	$\|\kappa A_1\|_{expt}$[a]	$\|nA_n/A_1\|$ Experiment	Lifshitz-Kosevich theory[b]	Magnetic-interaction theory[c]
Noninteraction case: $\alpha[111]$; $H(\alpha_1)=130.8$ kG, $H(\alpha_2)=150.0$ kG, $\omega/2\pi=135$ kHz, $T_D=0.00$°K					
α_2	3.5	1.3×10^{-2}	6.1×10^{-2}	6.1×10^{-2}	1.3×10^{-2}
	1.0	5.3×10^{-2}	7.3×10^{-1}	7.6×10^{-1}	5.3×10^{-2}
Noninteraction case: $\delta[111]$; $H(\delta_1)=117.0$ kG, $H(\delta_2)=141.0$ kG, $\omega/2\pi=135$ kHz, $T_D=0.00$°K					
δ_2	3.5	1.3×10^{-2}	2.5×10^{-2}	2.0×10^{-2}	1.3×10^{-2}
	1.0	7.6×10^{-2}	2.1×10^{-1}	3.6×10^{-1}	7.6×10^{-2}
Strong-fundamental case: $\beta[100]$; $H(\beta_1)=61.8$ kG, $H(\beta_2)=79.4$ kG, $H(\beta_3)=87.7$ kG, $\omega/2\pi=145$ kHz, $T_D=0.08$°K					
β_2	2.1	1.0×10^{-1}	1.0×10^{-1}	2.6×10^{-2}	1.0×10^{-1}
	1.0	6.1×10^{-1}	3.4×10^{-1}	2.6×10^{-1}	6.1×10^{-1}
β_3	2.1	1.3×10^{-1}	2.6×10^{-2}	2.2×10^{-3}	1.8×10^{-2}
	1.0	6.0×10^{-1}	1.7×10^{-1}	4.7×10^{-2}	4.0×10^{-1}

[a] The field dependence of the LK expression (2.4) was used to adjust the observed value of $|\kappa A_1|$.
[b] Values of g were assumed to be 5.6 for the β oscillations and 4.7 for the α and δ oscillations (see Table VII). Values of μ were taken from Table III, Col. 3.
[c] Calculated from the strong-fundamental solutions (2.9) and using the adjusted experimental values of $|\kappa A_1|$; knowledge of the g values is not required for the results in this column.

$|\kappa A_1|=\kappa(V/V')nI_0$, where I_0 is the (constant) amplitude of the current in the driving coil (in abamperes). Since the net amplification of the circuitry depends on which resonant frequency is used and which filters are in the circuit, it was necessary to calibrate each particular arrangement used in the actual experiments, and corrections were made for samples which were larger or smaller than the driving coil. Although this method of calibration should be reliable to better than 10%, there remains the possibility that the samples were not always centered within the pickup coil (in both a radial and an axial sense), and indeed the absolute amplitudes from different crystals were found to vary by as much as 50%. The measured values of $|\kappa A_1|$ for several oscillations are listed in Table VII, and in all cases $|\kappa A_1|$ is found to be less than unity at the high fields used in these experiments. Although the values of $|\kappa A_1|$ were actually measured at different field strengths, the field dependence of the LK expression (2.4) was used to adjust the amplitudes to a common field of 100 kG.

According to the results of our temperature-dependence studies, the fundamental amplitudes should be correctly given by the ideal LK theory. All but three of the parameters required to evaluate $|\kappa A_1|$ from (2.4) and (2.5) are known from this work, and values of F and the CF were taken from the recent results of Anderson, O'Sullivan, and Schirber (private communication; see Tables III and VII). Since nothing is known about the values of the remaining parameter g for the various orbits, the theoretical results are presented in Table VII in the form of the magnitude of the spin-splitting factor $\cos(\frac{1}{2}\pi g\mu)$ which would be required to give exact agreement with the experimental amplitudes. In most cases the magnitude of $\cos(\frac{1}{2}\pi g\mu)$ is fairly small so that the fluctuations of the observed amplitudes from sample to sample have little effect on the lowest value of the argument of the cosine factor. The g values inferred from the spin factor using the values of μ given in Table III (Col. 3) are found to differ appreciably from the free-electron value of 2.0023, as might be expected on account of the strong spin-orbit interaction in lead,[4] and it seems that the lowest common g factor for all orbits on the third-zone electron surface (monster) is about 5.6 whereas that for the second-zone hole surface is about 4.7. On the other hand, if we pick the lowest g values for $g>1.5$, the product $g\mu$ for *all* orbits on *both* sheets of the Fermi surface turns out to remarkably constant, varying between 2.78 and 2.93.

Values of the absolute amplitude $|4\pi dM_{a\pm b}/dB|$ for several combination terms were also measured and are given in Table VIII, and it can be seen that the amplitudes of some of these terms are almost as large as some of the fundamental amplitudes (Table VII). We have also calculated the values of the combination amplitudes expected from (2.12) using the experimental values of A_a and A_b, and these calculated amplitudes are found to agree nicely with those observed directly.

The observed strengths of several harmonics are presented relative to the fundamental amplitudes in Table IX, where they are compared with the harmonic content expected from the ideal Lifshitz-Kosevich theory on the one hand [Eq. (2.4)], and the strong-fundamental solutions of the magnetic-interaction theory on the other [Eq. (2.9)]. The harmonic content for the α and δ oscillations is found to agree well with the former, whereas that for the β oscillations is best explained by the latter (especially at the higher temperatures), and these findings are completely consistent with our deductions from the temperature-dependence results in Sec. IV A.

ACKNOWLEDGMENTS

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