INVESTIGATION OF THE FERMI SURFACE OF LEAD BY THE CYCLOTRON RESONANCE METHOD

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Cyclotron resonance has been investigated in the (100) and (011) planes in lead monocrystals. The anisotropy in the effective mass of the conduction electrons corresponding to the lowest cyclotron resonances has been studied. A comparison has been made and agreement found between the experimental results and a Fermi surface model based upon the free electron approximation. [4,5] The dimensions of the multiply-connected surface of the third zone have been determined.

CYCLOTRON resonance studies yield a variety of experimental data from which the correctness of one or another model for the Fermi surface of a metal can be verified. [1] Cyclotron resonance in lead has been investigated by Aubrey; [2] however, poor definition and the variety of possible interpretations of the spectra which he obtained rendered impossible a quantitative comparison between them and the Fermi surface model. Various reasons for failures of this sort are discussed in [1].

THE EXPERIMENT

The measurements were carried out, using a frequency-modulation technique, [3] on lead monocrystals, characterized by a resistance ratio $\rho(20°C)/\rho(4.2°K) = (0.6 - 1) \times 10^4$. The crystals had the form of rectangular slabs of dimensions $13 \times 6 \times 1$ mm, and were grown from a melt in demountable forms of polished quartz. Their natural surfaces were not subjected to further working. The directions of the crystal axes of the samples were monitored by x-ray methods to an accuracy of $\sim 1°$. Figure 1 shows the orientations of the axes of the two samples investigated, as well as the directions of the rf currents and the magnetic field, which was oriented in various directions in the plane of the sample. Certain questions regarding the experimental conditions—in particular, the orientation of the magnetic field—have been discussed in [1]. The measurements were made at a frequency of 9.47 kc in magnetic fields ranging from 0.8 to 7 kOe, at a sample temperature of 2°K.

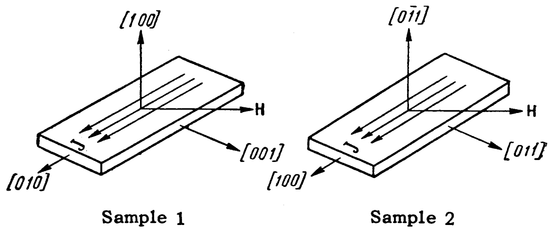

FIG. 1. Orientation of crystallographic axes for the two lead monocrystals studied. The arrows J indicate the direction of the rf currents. The magnetic field H lies in the plane of the sample.

A sample trace of the cyclotron resonance spectrum is presented in Fig. 2. From each such spectrum, obtained for successive rotations of the field through 2—3°, the ratio μ of the effective electronic mass m^* to the mass of a free electron m_e was determined from the formula

$$\mu = m^*/m_e = H_\omega^{-1}/(H_{n+1}^{-1} - H_n^{-1}), \qquad (1)$$

where H_ω is the field strength at electron paramagnetic resonance, while H_n is the field at the cyclotron resonance of order n.

The depth of the cyclotron resonances in each group, corresponding to a particular effective mass, depends fundamentally upon the direction of the magnetic field. Whatever the orientation of the field, however, the resonances in the group ζ, are of appreciably greater depth than the rest, as in the spectrum in Fig. 2.

The dependence of $\mu(\varphi)$ for the various cyclotron resonances upon the angle φ of the magnetic field relative to the crystallographic axes is displayed in the polar diagram in Fig. 3. This dia-

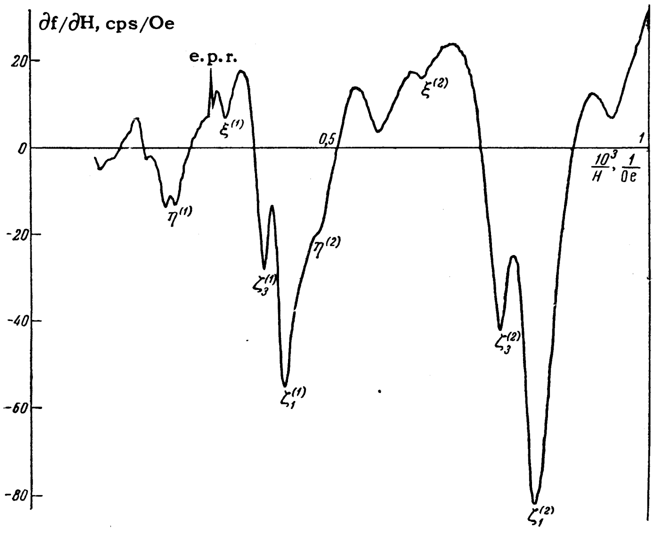

FIG. 2. Cyclotron resonance spectrum trace obtained for sample 1 (Fig. 1) with field rotated 3° from the [001] axis. The symbols adjacent to the resonances (minima) correspond to those for the effective masses in Fig. 3; the indices in parentheses represent the order of the resonances. (e.p.r.—electron paramagnetic resonance).

gram consists of two sections, joined along the [011] axis. The data obtained from experiments on sample 1 are presented in the lower portion, and those for sample 2, in the upper (see Fig. 1). The polar diagram thus represents a simple dihedral surface, unfolded along the [011] axis into a plane. Data on the effective masses could not be obtained in these experiments for $\mu \lesssim 0.3$, since they were carried out in magnetic fields exceeding ~ 1 kOe in the normal-state regions of the lead samples.

DISCUSSION OF THE RESULTS

In constructing a model for the Fermi surface one may make use of the concept of free electrons situated in a weak crystalline field. In accordance with this model, [4,5] the first Brillouin zone is filled; the second zone has a closed central hole surface. The third zone is filled near its edges; its energy surface, in the form in which it has been constructed by Gold, [4] is illustrated in Fig. 4. This surface is made up of identical tubes whose axes lie in the ⟨011⟩ directions.

Let us consider the possible character of the cyclotron resonances whose excitations are to be expected to derive from the form of the surface of the third zone (Fig. 4). Resonances may be observed for the extremal closed orbits lying in planes perpendicular to the magnetic field. Thus, for a field directed along the [001] axis, resonance should occur for the orbits ζ_1, ζ_2, ζ_3, ζ_4, ξ, and η, while the corresponding effective masses are determined by the well-known relation

$$2\pi m^* = \partial S(\varepsilon)/\partial\varepsilon, \tag{2}$$

where $S(\epsilon)$ is the cross-section area of the surface of constant energy ϵ in which the orbit lies.

Let us consider a cylindrical tube of the Fermi surface. For two cross sections of this tube—$S_0(\epsilon)$, perpendicular to its axis, and $S_\varphi(\epsilon)$, inclined at an angle φ —we have

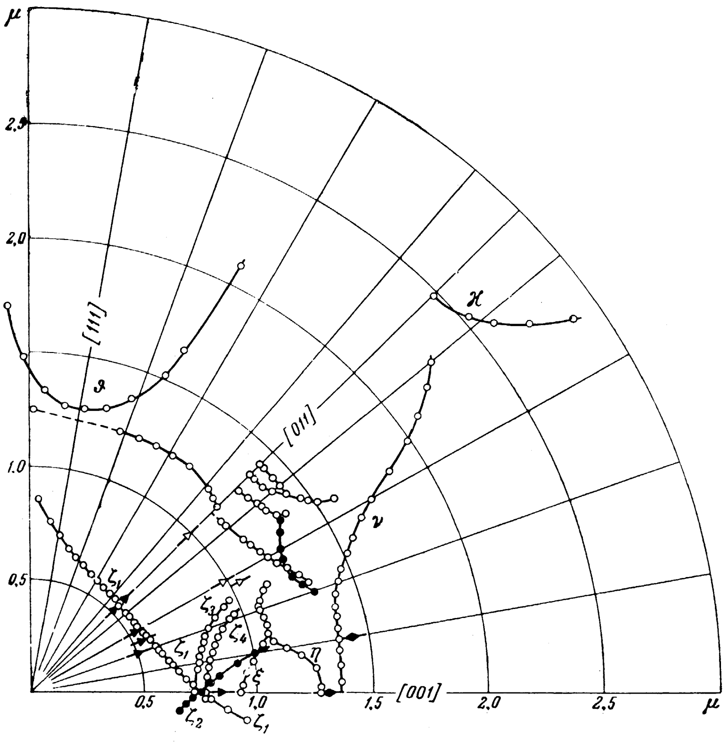

FIG. 3. Effective masses $\mu = m^*/m_e$ for the conduction electrons in lead in the following planes: (001)—lower half of diagram; (011)—upper half. The masses found by Gold[4] are indicated by the symbols: Δ—from the α-oscillations, ◆—from the β-oscillations, and ▲—from the γ-oscillations.

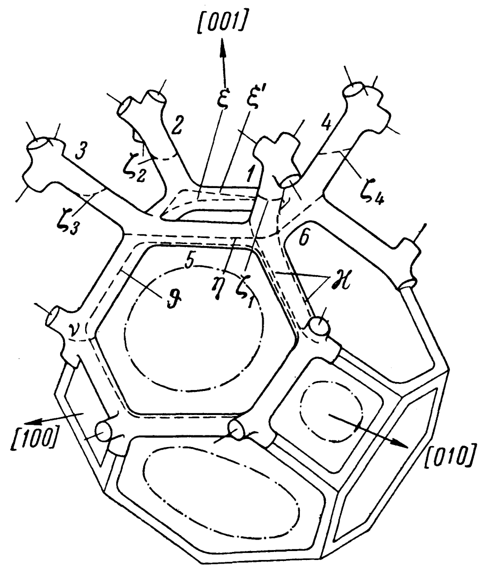

FIG. 4. Electron Fermi surface for the third zone. The dashed lines indicate the resonance orbits for the electrons: ζ_1, ζ_2, ζ_3, ζ_4, ξ, η, and ν – with the field parallel to the [001] axis; ϑ – with the field parallel to the [111] axis; $\varkappa$ – with the field parallel to the [011] axis. For clarity, the tubes have been reduced in diameter by approximately one-half; the dot-dash contours approximate the actual dimensions.

$$(S_\varphi(\varepsilon_1) - S_\varphi(\varepsilon_2))/(S_0(\varepsilon_1) - S_0(\varepsilon_2)) = 1/\cos\varphi, \quad (3)$$

where $\epsilon_2 \neq \epsilon_1$. From this, it follows that

$$m^*_\varphi/m^*_0 = (\cos\varphi)^{-1} = S_\varphi(\varepsilon)/S_0(\varepsilon), \quad (4)$$

i.e., the effective mass of the electrons in this case is proportional to the area enclosed by the orbit. For the actual energy surface, this relation is the more nearly fulfilled the less the departure of the surface from the cylindrical form.

Let us now investigate the variation of the orbits ζ_1, ζ_2, ζ_3, and ζ_4 as the field is rotated. Rotation of the field from the [001] axis to the [011] axis leads to an increase in the area enclosed by the orbits ζ_2, ζ_3, and ζ_4, and a decrease in that of the orbit ζ_1. The areas of the orbits ζ_3 and ζ_4 remain equal, increasing less rapidly than that of the orbit ζ_2. At the same time, the area of the orbit ζ_1 decreases, as the field is rotated through a small angle φ away from the [001] axis, at the same rate as the area of the orbit ζ_2 increases. It should be noted that for a possible error of $\sim 1°$ in the orientation of the sample, the areas of the orbits ζ_3 and ζ_4 will deviate slightly to either side of the mean value corresponding to exact orientation. The complete set of effective masses ζ_1, ζ_2, ζ_3, and ζ_4 is displayed in the diagram (Fig. 3); their behavior corresponds exactly to the pattern we have set forth.

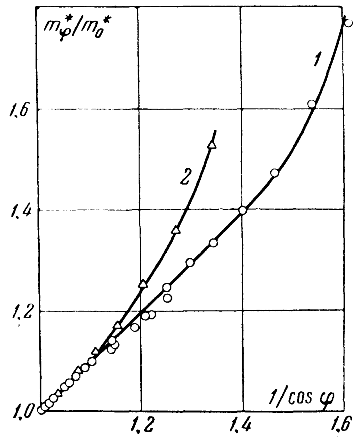

FIG. 5. Data concerning the form of the tubes making up the Fermi surface illustrated in Fig. 4.

With the aid of Eq. (4) we can determine the degree to which the form of the tubes making up the Fermi surface approximates the surface of a cylinder. Figure 5 shows the dependence of $m^*\varphi/m^*_0$ upon $(\cos\varphi)^{-1}$ for two crystal planes: (100) — curve 1, and (011) — curve 2. It is evident from the figure that, within experimental error, the tube is cylindrical over a length of the same order as its diameter. The curve passing through the experimental points lies at an inclination of 45° relative to the x axis; this confirms the supposition that the axis of the tube is parallel to the [011] axis. The difference between curves 1 and 2 bears witness to the fact that the cross-section of the tube is not circular. The nearly-cylindrical form of the tubes explains the relatively great depth of the cyclotron resonances in the orbits ζ_1, ζ_2, ζ_3, and ζ_4.

It is difficult to predict the way in which the effective masses corresponding to the orbits ξ and η should vary as the field is rotated from the [001] to the [011] axis. Over a range of a few degrees from the [001] axis, however, the area of the orbit ξ should increase, while that of the orbit η should decrease. Qualitatively, this corresponds to the behavior of the effective masses ξ and η (Fig. 3).

As is evident from Fig. 4, cyclotron resonance should occur in the orbit ϑ in the presence of a magnetic field along the [111] axis, and the corresponding effective mass should increase rapidly with deflection of the field. The cyclotron resonance found experimentally for the orbit ϑ is indicated in Fig. 3. Knowing the range of orientations of the magnetic field over which cyclotron resonance in the orbits ζ_1, ζ_2, ξ, and ϑ is observed, one can determine the mean diameter of the tubes (in units of $2\pi/a$, where a is the lattice constant). These data are presented in Table I.

Table I

Orbit	Plane of rotation of **H**	Computational formula*	Mean Diameter of tube in units of $2\pi/a$
ζ_1	(100)	$\cot 55° = d/(l-d)$	0.29
ζ_1	(011)	$\cot 42° = d/(l-d)$	0.37
ξ	(100)	$\tan 25° = d/(l\sqrt{2}-d)$	0.32
ϑ	(011)	$\tan 16° = d/l\sqrt{3l}$	0.34
ϑ and ζ		Eq. (5)	0.37
	Tube diameter, average of all values		0.34

*The tube length is $l = (2\pi/a)/\sqrt{2}$.

From Eq. (4) of the paper by I. Lifshitz, Azbel', and Kaganov,[6] it follows directly that

$$\oint v_{\perp}^{-1}\, dl = 2\pi m^{*}.$$

Assuming the orbits ϑ and ζ to be circular (for, respectively, **H** ∥ [111] and **H** ∥ [011]), and taking account of the fact that $v_{\perp}$ is constant and identical in both, since these orbits have a common point, we arrive at the conclusion that the corresponding effective masses must be proportional to the diameters of the orbits; i.e.,

$$\mu_{\vartheta}/\mu_{\zeta} = (\sqrt{3/2} - d)/d. \quad (5)$$

Substituting the values for the masses obtained from Fig. 3, we obtain $d = 0.37\,(2\pi/a)$.

The effective masses κ and ν are also indicated in Fig. 3. From their behavior it is possible to associate them with the orbits identified by the same symbols in Fig. 4.

Thus, in the present work, cyclotron resonances have clearly been observed for all of the extremal orbits lying in the multiply-connected Fermi surface of the third zone. (It was not possible to detect a resonance in a field **H** parallel to the [111] axis corresponding to the orbit surrounding that part of the surface about whose interior the orbit ϑ lies; this was due primarily to insufficient field strength in the magnet, since the corresponding effective mass would be of the order of 5.)

Certain other effective masses, corresponding to cyclotron resonances of lesser depth, are also plotted in the polar diagram of Fig. 3. They can be attributed to orbits lying on the closed Fermi surface of the second zone, and also to the possibly partially filled energy surface of the fourth zone.[4] The presently available data, however, do not permit this assignment to be made with adequate certainty.

Let us now compare some of the experimental results from the present work with the data obtained from investigation of the de Haas–van Alphen effect.[4] The value of $0.38\,(2\pi/a)$ for the diameter of the tubes of the Fermi surface (Fig. 4), found by Gold[4] from the area of the extremal cross section $S = 0.11\,(2\pi/a)^2$, is in good agreement with the value for the average diameter (Table I) found in the present work: $d_{av} = 0.34\,(2\pi/a)$; in this connection, it should be recalled that the tube has a noncircular cross-section (Fig. 5).

In view of the fact that cyclotron resonance and the de Haas–van Alphen effect are both associated with the motion of electrons about identical orbits, it is possible to compare the ranges in the directions of the magnetic field over which these phenomena are observed; the agreement is illustrated in Table II.

Table II

Cyclotron resonance data (see Fig. 2)		Data from magnetic susceptibility oscillations[4]	
Orbits	Angular range	Type of oscillation	Angular range
ζ_3, ζ_4	± 21°–25°	γ ([4], Fig. 2, curve 2)	± 19°
ζ_1, in (011) plane	± 42°	γ ([4], Fig. 4, curve 3)	± 42°

Values for the effective masses of certain groups of electrons, as determined by Gold[4] from the temperature dependence of the magnetic susceptibility oscillation amplitudes in lead, are plotted in Fig. 3. The mass values for the α-oscillations are close to the masses tentatively assigned to the second-zone Fermi surface. For the γ-oscillations, full agreement is obtained, within the limits of experimental error, with the ζ orbit data.

The masses for the β-oscillations indicate that they are to be attributed to the orbit ν (Fig. 3), and not to the orbit ξ, as Gold has done.[4] This latter choice necessitated a number of unsatifactory assumptions regarding the form of the tubes, inasmuch as the diameter of the orbit (which we take to be circular) as calculated from the β-oscillations is relatively large: $\sim 0.6\,(2\pi/a)$. At the same time, it is quite natural to assume the existence of increase in the magnitude of this dimension in the vicinity of the junctions in the surface, at each of which four tubes come together. The most convincing grounds for the proposed interpretation, however, lies in the circumstance that the Fermi surface illustrated in Fig. 4, possessing such enlargements, has open ⟨111⟩ directions, whose presence in the Fermi surface of lead has been established by galvanomagnetic studies.[7]*

Thus the experimental study of a number of the cyclotron resonances, including the deepest, observed in lead shows good agreement, both qualitative and quantitative, with the form of the Fermi surface for the third zone based upon the free-electron approximation.

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*The fact that the Fermi surface model for lead proposed by Gold[4] can have open <111> directions, was pointed out by R. Young in a letter to Alekseevskii and Gaĭdukov,[8] who kindly transmitted this information to the present authors.

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