INVESTIGATION OF THE FERMI SURFACE OF ALUMINUM BY THE METHOD OF QUANTUM OSCILLATIONS OF THE HIGH FREQUENCY SURFACE RESISTANCE

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Quantum oscillations of the surface resistance in a magnetic field are investigated in aluminum single crystals by means of an NMR spectrometer at a frequency of 5 Mc at fields up to 12 kG and at a temperature of 1.6°K. The anisotropy of the extremal cross sections of the Fermi surface of aluminum in the (100) and (110) planes is derived from measurements of the periods of oscillation as a function of the inverse field for various directions of the field. Several characteristic features of the shape of the Fermi surface of aluminum follow directly from the obtained experimental results. A complete analysis of all the experimental results has been carried out by a comparison of the Fermi surface of aluminum in zone III with Ashcroft's model.

INTRODUCTION

QUANTUM oscillations of the high frequency surface resistance of metals in a magnetic field were considered theoretically by Azbel'[1] and were subsequently observed in [2-4]. The study of quantum oscillations of the surface resistance makes it possible to determine the same characteristics of the electron spectrum in a metal as a study of the de Haas–van Alphen effect, that is, the areas of extremal cross sections of the Fermi surface and the effective mass of the electrons. However, certain features of the new effect, in particular, the polarization dependence of the high frequency currents, facilitate the analysis of the experimental results.

In comparison with the static Shubnikov–de Haas effect, the observation of quantum oscillations at high frequency has advantages associated with the strong inhomogeneity of the electromagnetic field inside the metal.

In the first place, to measure the static effect on good metals with high conductivity, one must prepare thin specimens of wires or films, which increases the resistance of the sample. In the high frequency case, the role of such a film is played by the skin effect.

In the second place, the value of the quantum contribution relative to the monotonic part of the resistance must be greater at high frequency for oscillations on anomalously small groups of electrons with a small effective mass, if the metal contains a fundamental group with $m^*/m_0 \sim 1$. The fact is that in a certain interval of fields the Larmor radius r of light electrons becomes smaller than the penetration depth, whereas $r > \delta$ for the remaining electrons. It is clear that in this case the relative contribution of the light electrons to the surface resistance must increase. In the present research, as the given estimates show, the quantum oscillations in aluminum were observed precisely in this field region.

The quantum oscillations of the magnetic susceptibility (the de Haas–van Alphen effect) were first observed in aluminum and studied by Gunnerson.[5] His results were interpreted by means of the Fermi surface for the third zone as constructed by Harrison[6] in the approximation of almost free electrons. This surface consists of tubes directed along the [110] axis and connected by thin necks. The existence of such tubes is confirmed not only by the experiments of Gunnerson but also by the subsequent cyclotron resonance measurements.[7,8] However, on the basis of all the published experimental results, it is impossible to state with complete assurance that these tubes are connected. Theoretically, there are many possibilities,[6,9] since the shape of the Fermi surface of aluminum at the places of connection of the tubes depends strongly on the value of the lattice potential and the boundary Fermi energy.

Thus, aluminum is of interest in that the general character of the connectedness of its Fermi surface is determined by small pieces of surface, which can be studied by the method of quantum oscillations in comparatively weak fields (up to 12 kG).

EXPERIMENTAL WORK

Specimens. The single-crystal samples were prepared from aluminum of purity characterized by the resistivity ratio

$$\rho_{4.2°K} / \rho_{300° K} = 10^{-4}.$$

Oriented single crystals of aluminum were grown from the melt in an assembled graphite mold in an atmosphere of CO_2. The method of obtaining oriented seeds and single crystals is similar to that described by Sharvin.[10] The single crystals obtained in this fashion had the shape of discs of 17.5 mm in diameter and 2 mm thick and served as samples for the investigation without any additional treatment.

The measurements were carried out on two specimens, whose surfaces were parallel to the crystallographic planes (100) and (110), respectively. The deviation of the orientation of the (100) specimen from the correct value was ~ 1°, and that of the (110) specimen ~ 2°. The orientation was determined by x-ray diffraction.

Scheme of measurement. For observation of quantum oscillations, an NMR spectrometer was used as before.[4] The principal sensitive element of the measuring apparatus is a "threshold" oscillator assembled according to the circuit of Pound and Knight.[11] The specimen of the metal under study is placed in the coil of the resonant circuit of the oscillator. All the measurements were carried out at a frequency of 5 Mc.

The method of measurement is based on the fact that the quality factor, and therefore the oscillation amplitude of generation, depends on the surface resistance of the specimen. Measurements were carried out by a modulation method. The magnetic field was modulated at 30 cps and with an amplitude from 10 to 100 Oe. The amplitude modulation of the generator, which arises as the result of the dependence R(H), is proportional to dR/dH. Recording of the signal was with an electronic automatic plotting potentiometer.

The details of the block diagram of the measurement apparatus do not differ from that usually employed in NMR spectrometers, with the exception of the system for stabilization and regulation of the amplitude, and consequently the sensitivity of the threshold oscillator. Changes in this part of the circuit were necessary because of the monotonic variation of the resistance of the metal in the magnetic field. When recording oscillations over a wide range of fields, the oscillation conditions change appreciably; therefore, to maintain the required oscillation amplitude, strong feedback is necessary. In the feedback circuit, a standard amplifier UM-109 is used with a vibrator inverter, to the input of which was applied the difference between the dc voltage proportional to the oscillation amplitude and the reference voltage.

The output of the amplifier was connected to an RP-5 relay which controlled the bias voltage on the grid of the second triode in the Pound-Knight circuit. By changing the reference voltage, one could regulate the sensitivity of the circuit. Calibration of the sensitivity was carried out periodically during the course of the experiment. For this purpose, a resistance of 300 kΩ was connected in parallel with the circuit by means of a relay, fed from the 50 cps line.

Measurement of the magnetic field. The magnetic field was measured by means of a germanium Hall pickup, attached to one of the poles of the magnet. For improvement of the linearity of the Hall pickup, it was placed at an angle to the magnetic field so that $v_x = f(H_{eff}) = f(H \cos \varphi)$, that is, the initial linear part of the characteristic of the pickup was used. Thus the scale in fields up to 12 kG was uniform within 3%. Calibration of the magnetic field by proton resonance was repeated periodically during the experiment. The signal from the Hall pickup was fed to the x coordinate of an automatic plotter, the circuitry of which was such that the location of the carriage was proportional to the reciprocal of the applied voltage and consequently to the intensity of the magnetic field.

The cryostat. Measurements were carried out at a specimen temperature of 1.6°K. The specimen, together with the induction coil attached to it, was vacuum isolated from the helium bath by a copper screen. Heat exchange was carried out by means of gaseous helium.

The change in the orientation of the specimen in the magnetic field was achieved by rotation of the entire cryostat; in this case the magnetic field remained always parallel to the surface of the specimen.

Change in the polarization of the high frequency currents during the time of the experiment was brought about by rotation of the coil by means of a rod introduced into the cover of the apparatus. The angle of rotation was read on a graduated dial accurate to ± 3°.

EXPERIMENTAL RESULTS AND DISCUSSION

In the range of fields studied (up to 12 kG) all the observed oscillations, as in the work of Gunnerson,[5] could be clearly divided into two groups: short-period (γ oscillations) and long-period (α

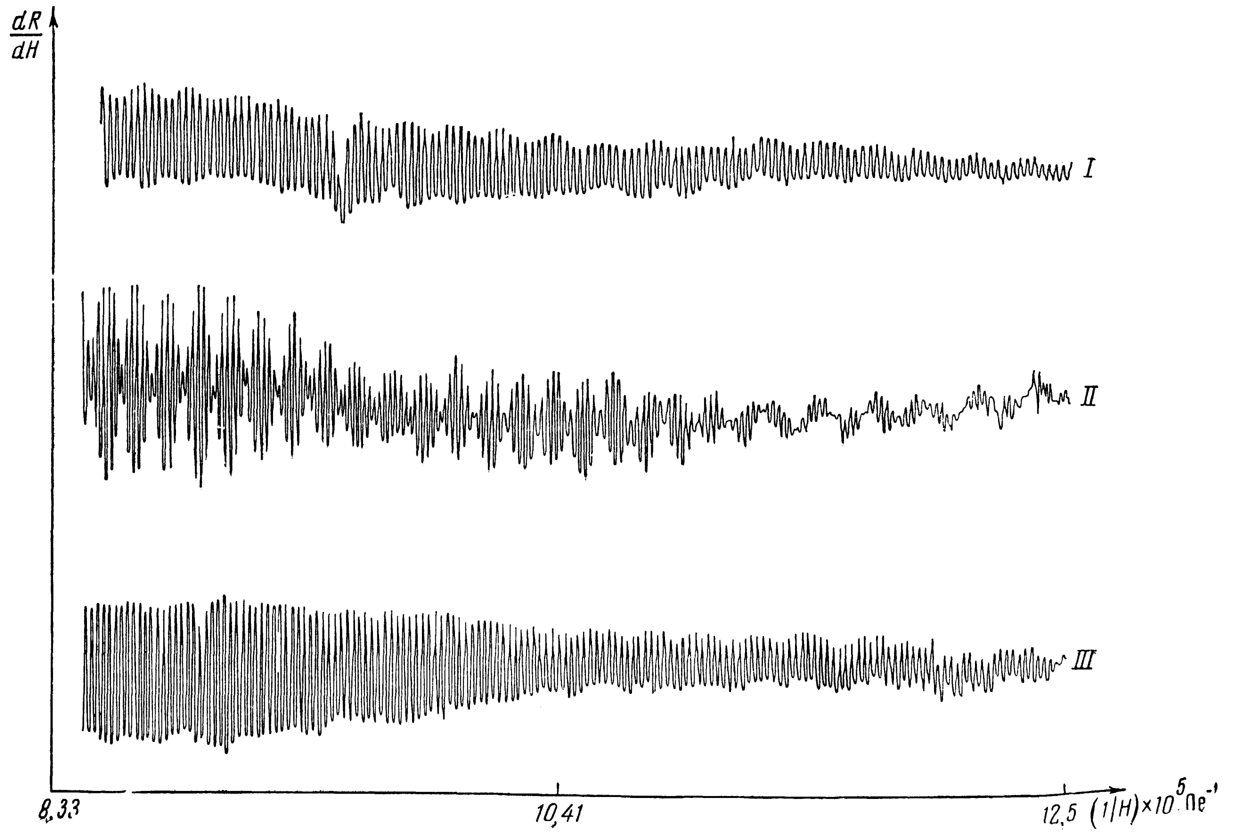


FIG. 1. Records of short-period quantum oscillations of the surface resistance on an aluminum single crystal with surface parallel to the (100) crystallographic plane. The directions of the magnetic field and the high-frequency currents, calculated from the [100] axis in this plane for the different curves are as follows: for the curve I–$\alpha_H = 11°$, $\alpha_j = -45°$; II–$\alpha_H = 11°$, $\alpha_j = +45°$; III–$\alpha_H = 0°$, $\alpha_j = +45°$.

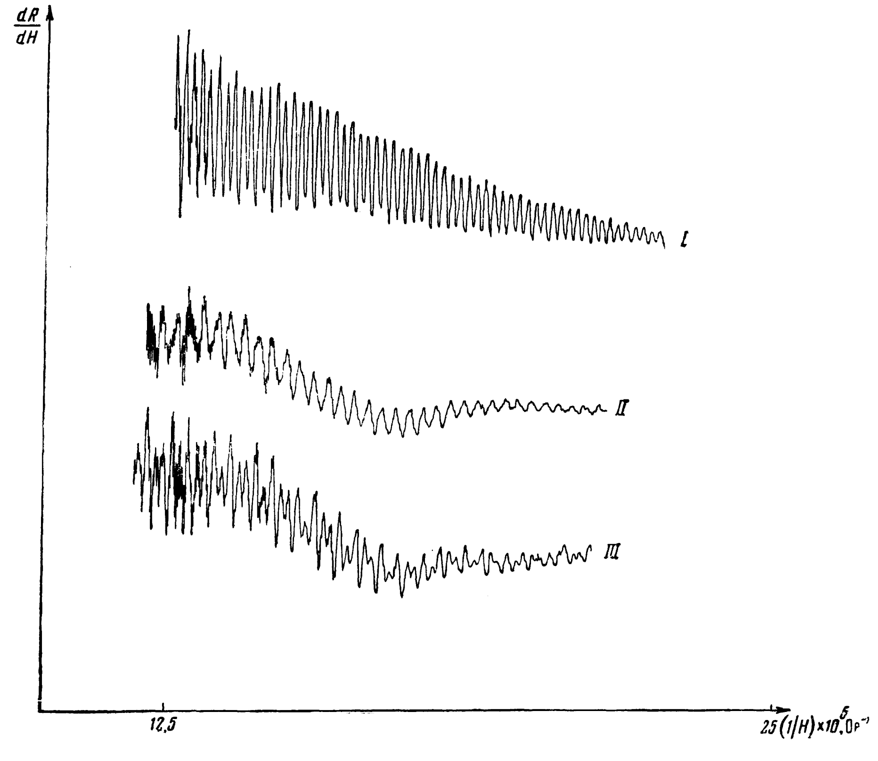


FIG. 2. Records of the long-period quantum oscillations of the surface resistance in an aluminum single crystal, whose surface is parallel to the (110) crystallographic planes (curve I) and the (100) planes (curves II and III). In the region of 12.5×10^{-5} Oe^{-1} (8 kOe) short-period oscillations appear. The directions of the magnetic field relative to the [100] axis are the following: I–$\alpha_H = 15°$, II–$\alpha_H = 2°$, III–$\alpha_H = 0°$.

and β oscillations), with periods $(1.5-3.5) \times 10^{-7}$ Oe^{-1} and $(20-40) \times 10^{-7}$ Oe^{-1}. The long-period oscillations were observed in fields of 4–5 kG, and the short-period ones above 8 kG. Typical experimental recordings are shown in Figs. 1 and 2.

From the periods of the quantum oscillations

$$\Delta(1/H) = eh/cS_{ext}$$

it is possible to determine directly the extremal sections through the Fermi surface by a plane perpendicular to the direction of the magnetic field in the crystal. However, if there is more than one extremal section for a given direction of the magnetic field, then the superposition of the corresponding oscillations makes the determination of the periods of the individual components more difficult. To make the analysis of the experimental data easier, several procedures were used, such as plotting the oscillations against the inverse field, the suppression of the short-period oscillations by large-amplitude modulation of the magnetic field, and suppression of one of the components by selection of the polarization of the high frequency currents.

The results of investigations of the two crystallographic planes (100) and (110) are plotted in Figs. 3–6 in polar coordinates. The radius vector in these diagrams determines the value of the extremal section of the Fermi surface and the corresponding direction of the magnetic field.

We shall discuss the results for the two groups of oscillations separately.

Short period oscillations. Figures 3 and 4 show, in addition to the experimental points, straight lines which represent the angular dependence of the cross sectional areas of six identical cylinders, the generating lines of which are parallel to the [110] axis. The minimum experimentally observed cross section for H ‖ [110] is taken as the starting point in the construction of the lines. The shape of the cross sections of the cylinders can be arbitrary. The cross sections are produced by a plane perpendicular to the direction of the magnetic field.

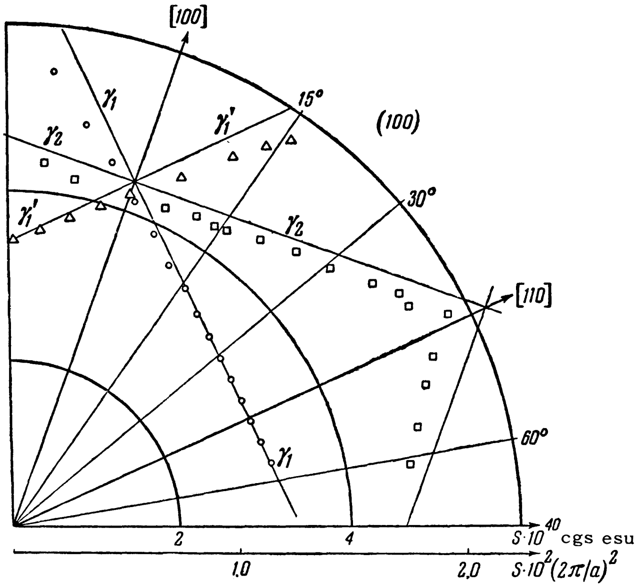


FIG. 3. Areas of the extremal cross sections of the Fermi surface of aluminum, determined from the periods of the short-period quantum oscillations for different directions of the magnetic field in the (100) plane.

The equation of the straight lines is

$$S_n = S_0^{(n)} / \cos(\varphi - \varphi_n);$$

$S_0^{(n)}$ is the maximum cross section of the cylinder for a magnetic field parallel to the given plane, and φ_n is the direction of the magnetic field corresponding to it.

Comparison of the experimental results with the straight lines thus constructed makes it possible to draw the following conclusions.

1. The parts of the Fermi surface of aluminum on which the short-period oscillations are observed are tubes which are cylindrical in the central region and directed along the [110] axis. In addition to the anisotropy of the cross sections, proof of

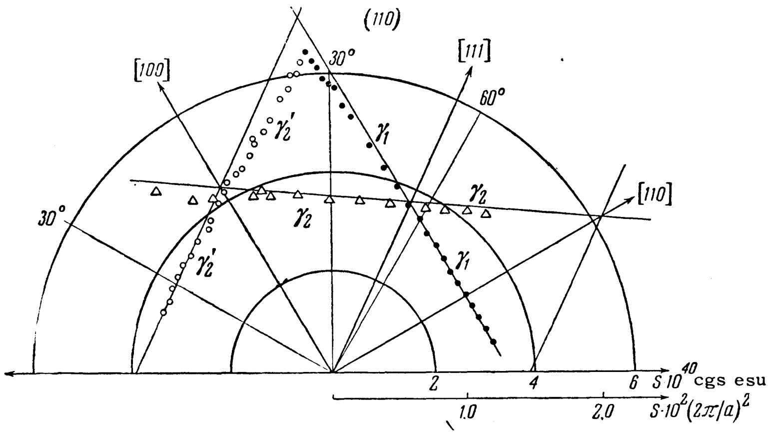


FIG. 4. Areas of the extremal cross sections of the Fermi surface of aluminum, determined from the periods of the short-period quantum oscillations for different directions of the magnetic field in the (110) plane.

this assertion is the character of the dependence of the amplitude of oscillations on the polarization of the high frequency currents. If the currents are directed along the tube, then the oscillations from it are entirely suppressed for all directions of the magnetic field. This is explained by the fact that the vector velocity of the electron $v = \partial\epsilon/\partial p$ is directed along the normal to the Fermi surface, and in the case of a cylindrical surface its projection on a direction along the cylinder is equal to zero; consequently, there is no contribution to the current from this part of the Fermi surface.

The effect of polarization of currents is demonstrated by the upper two curves in Fig. 1, which have been obtained for the same direction of the magnetic field. For these directions there are three branches (see Fig. 3). On curve 1 are plotted the oscillations from the lowest and most intense branch; for another polarization, these oscillations are suppressed and beats of oscillations from the other two branches are seen (curve II).

2. The tubes grow narrower toward their ends. This is seen by the deviation of the points from the straight lines.

3. The decrease of the transverse dimension of the tube along the direction toward its ends takes place more rapidly in the (100) plane than in the (110) plane. This conclusion follows from a comparison of branches γ_1 and γ_1' in Fig. 3, and the branch γ_1 in Fig. 4. The corresponding tubes are parallel to the crystallographic planes studied. Deviation of the experimental points from straight lines for a field inclined $\pm 60°$ to the axis of the tube are almost twice as great in the (100) plane as in the (110) plane and amount to about 10% of the value of the extremal cross section in this direction.

The error in the period of oscillations, without account of systematic errors connected, for example, with the nonlinearity of the Hall pickup, amounts to a fraction of 1% for a direction of the field along the tube and rises with increasing distance from it to about $\sim 3\%$ for the limiting points. The increase in the measurement error results from the fall in the amplitude of oscillations, as a consequence of which the interval of fields in which they can be seen becomes narrower.

The absolute accuracy of the periods and cross sections amounts on the average to 2%.

It should be emphasized that the conclusions given above follow directly and uniquely from the experimental results, and do not depend on any sort of definite model of the Fermi surface. In addition to the described measurements of the extremal sections, the effective mass was obtained from the temperature dependence of the amplitude of oscillations for the direction H $\parallel$ [110], and is equal to

$$m^* / m_0 = 0.133 \pm 0.01.$$

Measurements of the cyclotron resonance[8] for masses that are similar in anisotropy give a value $m^*/m_0 = 0.14$.

The extremal cross section in the central part of the tube for H $\parallel$ [110] is equal to $1.15 \times 10^{-2} \times (2\pi/a)^2$, which gives for the average diameter $d_{av} \approx 1.21 \times 10^{-1} (2\pi/a)$. Since the oscillations were tracked in the range of angles $\pm 60°$ from this direction, then the length of the middle part of the tube (from the γ oscillations) is equal to $\sim 2.1 \times 10^{-1} (2\pi/a)$, or about one third of the total length of the tube.

Long-period oscillations. Long-period oscillations can be divided into two groups, according to the character of the anisotropy and according to the intensity. The corresponding branches are denoted by the letters α and β in Figs. 5 and 6. The α oscillations are more intense as a rule and more isotropic than the β oscillations. Measurements of the periods of the α oscillations especially in the (100) plane, are carried out in most cases by beats with the β oscillations (curve III in Fig. 2). These measurements are accurate to about 5%. The comparatively low accuracy of the measurements is explained by two reasons: in the

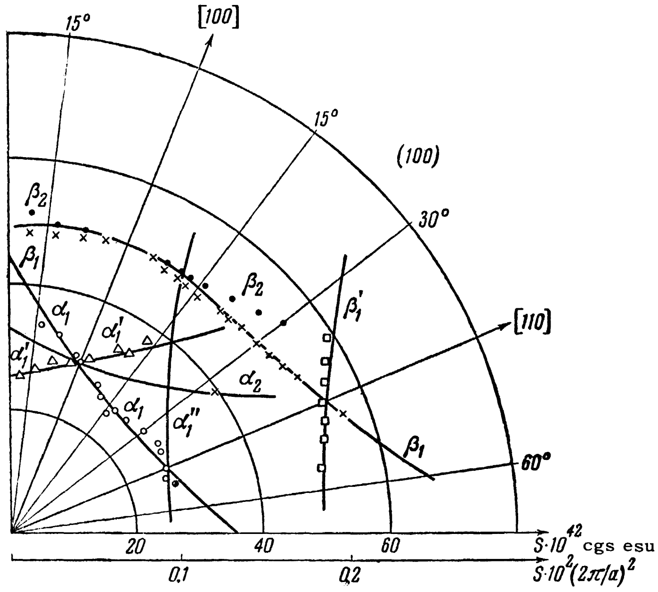


FIG. 5. Areas of the extremal cross sections of the Fermi surface of aluminum, determined from the periods of the long-period quantum oscillations for different directions of the magnetic field in the (100) plane.

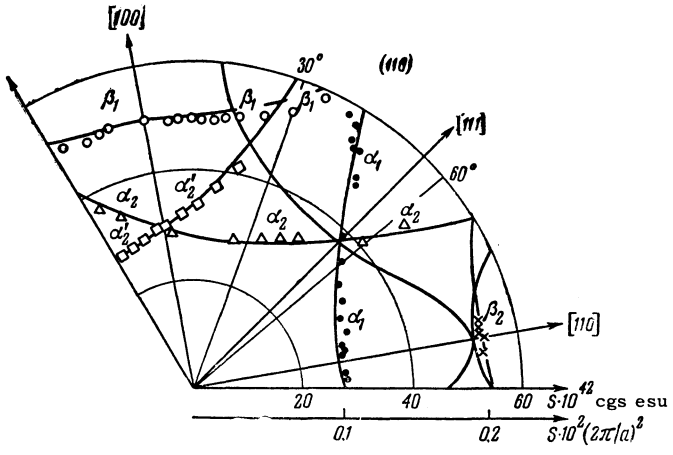


FIG. 6. Areas of the extremal cross sections of the Fermi surface of aluminum, determined from the periods of the long-period quantum oscillations for different directions of the magnetic field in the (100) plane.

first place, the $\sim 1:2$ ratio of periods is unsuitable from the view point of interpretation of the beat picture; second, these beats are usually clearly evident only in narrow field intervals. The accuracy of measurement of the periods of the β oscillations was approximately 1%.

By comparing Figs. 3 and 4 with Figs. 5 and 6, it is not difficult to ascertain that the anisotropies of the α and β oscillations are quite similar. Therefore it is natural to suppose that the α oscillations take place from orbits which lie in narrow necks on the ends of the tubes in zone III (Fig. 7).

It is very significant that the suppression of the individual branches of the α oscillations takes place approximately in the same polarizations of high frequency currents as for the corresponding branches of the γ oscillations. Thus, for example, in the (110) plane, one can separately observe the beats of α_2 and α_2' with β_1, by setting the polarization of the currents at an angle of approximately $\pm 35°$ from [100] axis.

It is seen in Fig. 6 that the deviation of the points of the α branch from straight lines is opposite in sign to that of the γ oscillations, which evidently agrees with the character of the curvature of the surface of the necks on the ends of the tubes.

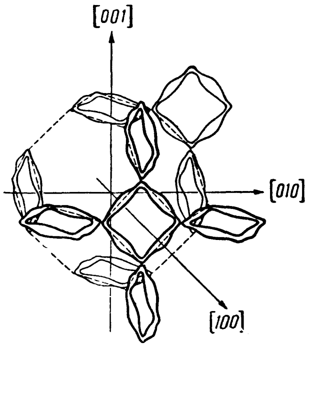


FIG. 7. Fermi surface of aluminum in zone III.

On the basis of the data of Figs. 5 and 6, it can also be concluded that the minimal cross section on the neck does not correspond to the field direction along [110]. The length of the neck amounts to approximately 5% of the length of the tube.

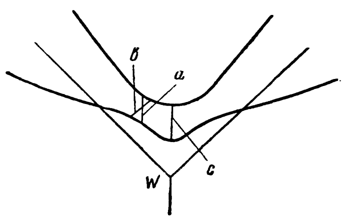
The Fermi surface of aluminum in zone III. Further interpretation of the results is carried out by comparison with the model of the Fermi surface of aluminum proposed by Ashcroft.[9]

The principal difference between this model and the well known model of Harrison[6] amounts to the fact that the tubes in zone III are connected not by fours but pairwise, so that the surface breaks up into isolated square "rings," which consist of four tubes each (Fig. 7). The qualitative interpretation of the γ and α oscillations is identical in either model if we refer the γ oscillations to orbits on the middle parts of the tubes and the α to orbits on the thin passages at the ends of the tubes. The advantage of the new model is the possibility of attaining with its help a convincing interpretation of the results on β oscillations.

In Fig. 8 (taken from [9]) is shown the section of part of the "ring" by the (100) plane through the point of symmetry W, and the orbits corresponding to long-period oscillations: α oscillations come from orbits a and b and β oscillations from orbits c on the thicker junctions of the tubes.

Theoretically calculated curves for the angular dependence of the periods of low frequency oscillations in the (100) plane were given in [9]. These curves, after the recalculation into sections, are plotted in Fig. 5, the experimental points lie very close to these curves, the divergence in most cases not exceeding the error of measurement.[1)] The branch β_2 on (100) plane is missing from the

FIG. 8. Part of the surface shown in Fig. 7. Orbits are shown which correspond to long-period oscillations.



[1)]Doctor Ashcroft was very kind and in answer to my request sent the curves calculated by him for the (110) plane. These curves are plotted in Fig. 6. Doctor Ashcroft also related that he considered also one branch for orbits of type c on the (100) plane, which correspond to the branches β_2 in Fig. 5.

theoretical curves,[9] but its presence follows from qualitative consideration of the model. In fact, if the field is directed precisely along [100], then the β oscillations will be seen from the orbit c on the rings whose planes are parallel to the field. There are two types of such rings: the plane of one coincides with the plane of the magnetic field, the other is perpendicular to this plane (Fig. 7). If the magnetic field is inclined to the [100] axis in the (100) plane, the corresponding branches (β_1 and β_2) should split. Since both branches should be symmetric relative to [100], for this direction of the magnetic field they should not intersect but should be tangent to one another. It is seen in the diagram of Fig. 5 how smoothly the points of the branches β_1 and β_2 diverge. The branch β_2 terminates without reaching the [110] axis (Fig. 5); therefore it must refer to orbits c on the ring which lies in the plane of rotation of the field.

The same orbits c are equivalent if the field is rotated in the (110) plane (branch β_1, Fig. 6).

The weak anisotropy of the periods of the β oscillations is explained by the fact that the shape of the thickened junction of the tubes is apparently close to spherical. Thus all of the special features of the anisotropy of the periods of the β oscillations agree with the model of [9].

From the foregoing discussion we see that the suggested identification of the electron orbits according to the model[9] (Fig. 7) for the three groups of oscillations observed in the present work makes it possible to interpret all the experimental results (Figs. 3–6). The completeness and accuracy of the results, and the excellent agreement with the theoretical curves,[9] provide convincing arguments in support of this model.

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