CYCLOTRON RESONANCE AND QUANTUM OSCILLATIONS OF THE SURFACE RESISTANCE OF BISMUTH

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Cyclotron resonance and quantum oscillations of the surface resistance of single crystals of bismuth are simultaneously investigated at a frequency 9.5×10^9 cps and temperature 1.7°K. Effective electron and hole masses are measured in two crystallographic planes and the cross sectional areas of the electron Fermi surface along directions close to the binary axis are determined. Some accurate and detailed quantitative characteristics of the Fermi surface of bismuth are obtained.

THE new experiments on the Fermi surface of bismuth were prompted by the recent theoretical study of Abrikosov and Fal'kovskiĭ [1], who demonstrated the possibility of a complete determination of the energy spectrum of the current carriers in bismuth from the experimental data.

The main experimental investigations of the Fermi surface of bismuth, using the de Haas–van Alphen effect, were made by Shoenberg [2–4], who proposed a detailed model of three ellipsoids for its electronic part. This model apparently agrees with the experimental results [2–12] within the limits of measurement error [1)]. The present work makes it possible to make more precise many characteristics of the electronic Fermi surface of bismuth.

Much less is known concerning the hole part of the Fermi surface of bismuth. Galt et al [9], in a study of cyclotron resonance in a circularly polarized high frequency field, observed resonance in the holes and measured their effective masses in two crystallographic directions. Brandt [10] suggests that the short-duration oscillations in the magnetic susceptibility of bismuth which he noted pertain to the hole surface. The form of the hole surface is represented in first approximation as an ellipsoid of revolution, the major axis of which is parallel to the trigonal axis of the bismuth. Thus, as the result of a large number of investigations of the electric and magnetic properties of bismuth it is known that it has closed electron and hole Fermi surfaces, the main characteristics of which have been determined. However, for the new theory of [1] it is necessary to investigate quantitatively in detail the Fermi surface, and in particular to ascertain the degree to which the quadratic dispersion law is valid. The purpose of the present research was to obtain accurate quantitative characteristics of the Fermi surface of bismuth.

EXPERIMENT

The experiments were carried out by the frequency-modulation method [13]: the logarithmic derivative of the surface reactance of bismuth was measured at a frequency 9.5 Gc as a function of the reciprocal of the intensity of the constant magnetic field, applied to the specimen parallel to its plane surface. The specimens were single crystals of bismuth in the form of discs 18 mm in diameter and 1.5 mm thick. The single crystals were grown from a melt in a dismountable glass mold, and their surfaces were not treated further. The original material was bismuth characterized by a ratio of the room-temperature to helium temperature resistance on the order of 100.

Four specimens were investigated: two had surfaces coinciding with the basal plane of the crystal, perpendicular to the trigonal axis (C_3), while two others had a surface containing the trigonal and binary (C_2) axes of the bismuth crystal.

The specimen was placed in a round strip resonator [13] under a resonating strip, so that the high frequency current flowed on one side of the crystal in a straight line. Rotation of the spe-

[1)]Incidentally, according to Weiner[8] and Lax[22], there are grounds for assuming that the electron dispersion law is not quadratic and the electron surface is not ellipsoidal.

cimen in the cavity made it possible to change the direction of the high frequency current in the specimen, that is, the polarization of high frequency field relative to the crystallographic directions of the specimen. It was thus possible to choose the experimental conditions that were optimal for the observation of cyclotron resonance[14,15]. The constant magnetic field could be rotated in the plane of the specimen. The magnetic field was maintained parallel to the specimen plane with accuracy ~ 5′[15].

A sample record of cyclotron resonances is shown in Fig. 1; each curve shows clearly a series of equidistant resonant peaks, which shift with rotating magnetic field H. In all the experiments the signal was so large that the sensitivity of the apparatus[13] had to be reduced by approximately two orders of magnitude. In fields larger than ~ 700 Oe, no cyclotron resonances were observed.

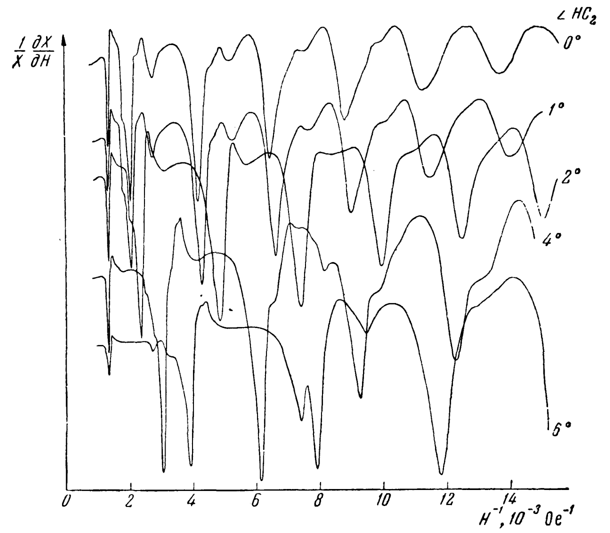

FIG. 1. Record of cyclotron resonances in a single crystal of bismuth, whose surface coincides with its basal plane. The series of resonances, which shifted rapidly with varying angle between the magnetic field and the binary crystal axis ($\angle HC_2$), pertains to the electrons of the Shoenberg model.[2,3]

In fields larger than ~ 500 Oe, quantum oscillations of the surface resistance of bismuth were observed. This effect was not yet investigated experimentally[2)], and mention of its observation was made in the paper by Aubrey[12]. A sample record of the quantum oscillations is shown in Fig. 2. The region where the quantum oscillations were observed was limited on the side of the weak fields

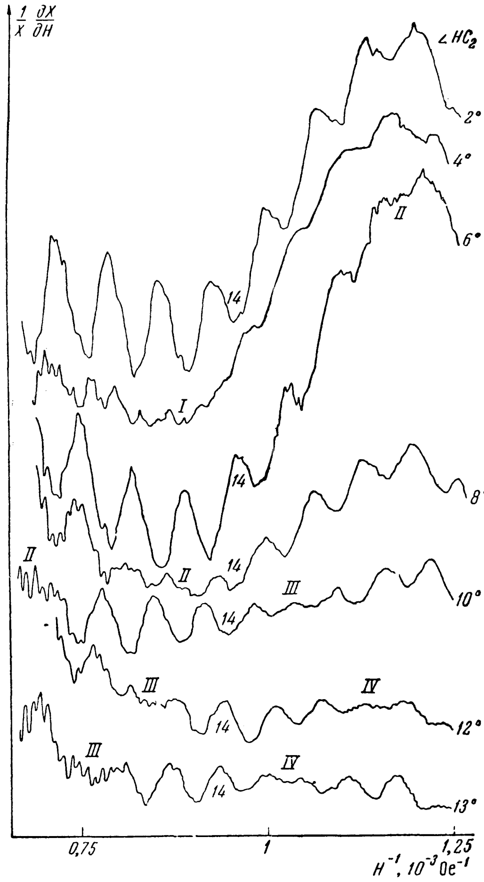

FIG. 2. Record of the quantum oscillations of the surface impedance of a single crystal of bismuth, with a surface whose plane coincides with its basal plane. On the right of the curves are marked the angles between the magnetic field and the binary axis (during the course of the experiments, records were taken usually in smaller angle intervals); 14 – number of the oscillations, counting from $H^{-1} = 0$; the Roman numerals denote the "fadings" of the beats. The rise in the right halves of the upper curves corresponds to the edge of the cyclotron-resonance peak.

by their reduced amplitude and the appearance of deep cyclotron resonances, while on the strong-field side it was limited by the appearance of rapidly growing oscillations with smaller period (they are seen on the left ends of the curves of Fig. 2). A separate article[17] will be devoted to a study of these oscillations of the surface impedance of bismuth.

RESULTS OF INVESTIGATION OF CYCLOTRON RESONANCE

The investigations of cyclotron resonance were aimed at measuring the effective masses of the electrons and holes, and also at an elucidation of those singularities in their anisotropy which can serve for a determination of the form of the Fermi surface of bismuth.

2) Quantum oscillations of the surface impedance of a metal under anomalous skin effect conditions were considered theoretically by Azbel'.[16]

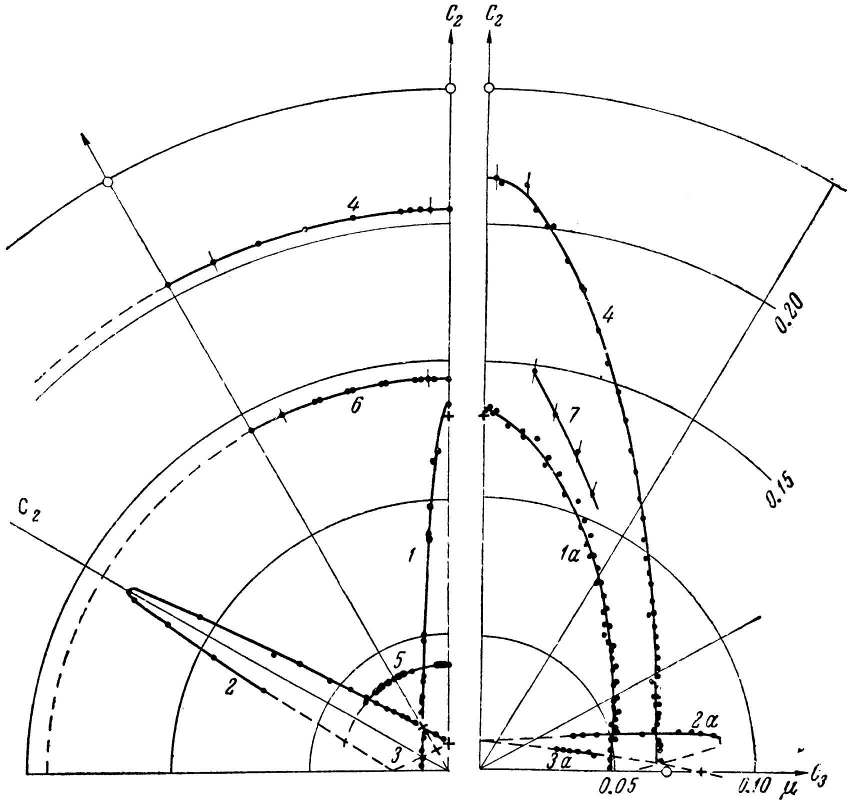

FIG. 3. Polar diagram of the dependence of the effective masses μ on the direction of the magnetic field applied to a single crystal of bismuth. The left quadrant represents the basal crystallographic plane, and the right quadrant the plane containing the binary axis C_2 and the trigonal axis C_3. The dashed curves are continuations of the experimental curves made in accordance with symmetry requirements. The values of the electron effective masses given by [9] are marked with crosses, the values of the holes are marked with circles (henceforth, the error bars near some of the experimental points indicate the accuracy of the measurement of the given mass). The figures designating the curves are explained in the text.

The effective masses were calculated from the formula[14]

$$\mu = \frac{m^*}{m_e} = \frac{e}{m_e c\omega \Delta_1 H^{-1}},$$

where $\Delta_1 H^{-1}$ is the period of the cyclotron resonances of the given mass, determined from experimental records similar to those shown in Fig. 1 (the remaining symbols are standard). Variation of $\Delta_1 H^{-1}$ by rotating the field in the plane of the specimen makes it possible to study the anisotropy of the effective masses, which manifests itself clearly on Fig. 1. Results of the measurements are shown on the polar diagram in Fig. 3; part of curve 2 is plotted on the basis of data from Fig. 1, pertaining to a sharply anisotropic mass.

Electronic Fermi surface. Curves 1 and 2, drawn through the experimental points of Fig. 3, are quite close to ellipses. Therefore, assuming the quadratic dispersion law to be approximately correct, we must conclude that the corresponding resonances occur on ellipsoidal Fermi surfaces. Inasmuch as the effective mass should in this case be proportional to the area of the extremal cross section of the ellipsoid, it is obvious that the major axes of the ellipsoids are perpendicular to the directions of the binary axes C_2, and the Fermi surface contains three such ellipsoids, separately located in the reciprocal-lattice space of the bismuth[3)]. The ratio of the major to the minor axes of the ellipsoid can be determined as the ratio of the maximum and minimum effective masses, and amounts to 14.8 (we have in mind here the ellipsoid minor axis perpendicular to the trigonal axis of the bismuth crystal).

From an examination of the curves 1a, 2a, and 3a on Fig. 3 we can conclude that the ellipsoids are located inclined to the basal plane, but a reliable determination of the angle of inclination from this data is difficult because of the inaccurate orientation of the surface of the given specimen (were the orientation accurate, curves 1a, 2a, and 3a would intersect at one point of the C_3 axis). The determination of this angle will therefore be made in the course of the analysis of the quantum-oscillation data (see below).

Figure 4 shows, on an enlarged scale, the behavior of the effective masses under consideration near their minimal values. The fact that the experimental curves fit well the lines described by equations of the type

$$\mu(\varphi) = \mu(30°)/\cos(\varphi - 30°)$$

3)From symmetry considerations,[1,18] the Brillouin zone of bismuth contains three pairs of "halves" of ellipsoids adjacent to the cell edges.

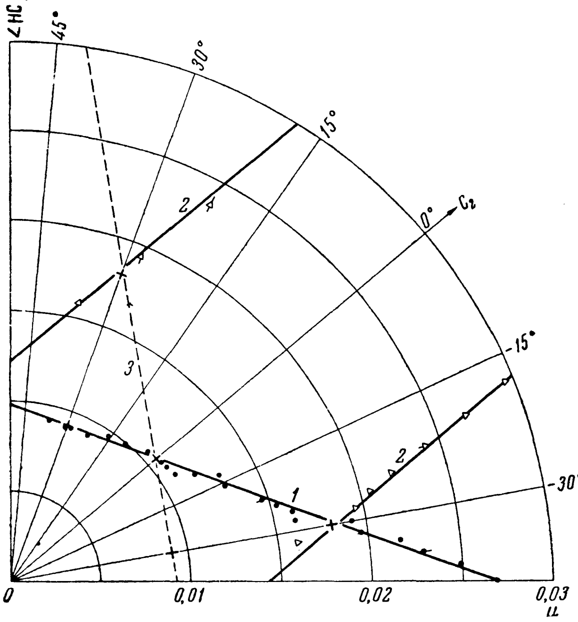

FIG. 4. Polar diagram of the dependence of the electron effective masses μ in the low-mass region on the direction of the magnetic field applied to a single crystal of bismuth in its basal place. The dashed curve 3 – mirror image of the experimental line 1, drawn in accordance with the symmetry requirements. Crosses – results of Galt et al.[9] The numbers of the experimental lines correspond to the numbers on Fig. 3.

(the equation of the line 1 on Fig. 4) indicates that the central parts of the ellipsoids cannot be distinguished, within the limits of the experimental accuracy, from cylinders with axes perpendicular to these lines[19]. This deduction will be used later on in conjunction with the results of the investigations of the quantum oscillations.

The characteristics obtained above for the three ellipsoids of the Fermi surface of bismuth are in full qualitative agreement with the model proposed by Shoenberg[2,3] for its electronic part.

Let us compare now some of the quantitative results of the described experiments with the available literature data. Figures 3 and 4 show the values of the electron effective masses, measured by Galt et al[9] for rational directions of the magnetic field; the very good agreement is obvious. The comparison with Aubrey's results[12] looks worse, for the discrepancies reach ~ 15 per cent. It is possible that the reason for the discrepancies lies in the low accuracy of Aubrey's measurements [12], which can be gauged from the experimental curves given in his paper. A comparison of the effective masses measured by the cyclotron resonance method with the values obtained from an investigation of the de Haas–van Alphen effect[4,8,11] is not illustrative for the present investigation, since the second method is much less accurate.

Let us consider now the ratio of the major and minor (perpendicular to C_3) axes of the electronic ellipsoid, as calculated from results of various investigations (see the table). In the case of investigations of the de Haas–van Alphen effect, this quantity was defined as the ratio of the sections of the Fermi surface, obtained for corresponding directions of the magnetic field (a correction for the orientation of the specimen was introduced where necessary). In the case of investigations of cyclotron resonance, the ratio chosen was that of the corresponding effective masses, which holds true only if the quadratic dispersion law is valid for the electrons. As can be seen from the table, the spread within the two groups of numbers pertaining to these two methods is ~ 20 and ~ 10 per cent, respectively. However, the difference between the averages of the groups reaches ~ 40 per cent. On the other hand, if we take in each group the latest results, which are the most reliable, the difference comprises ~ 60 per cent and is far beyond the limits of the measurement errors. It follows from this fact that generally speaking the quadratic dispersion law does not hold for the bismuth electrons (this makes the very designation "ellipsoid" for the electronic Fermi surface somewhat arbitrary). Later on, in the analysis of the investigations of the quantum oscillations, we shall consider the question of the region in which the quadratic dispersion law can be regarded as satisfied within the limits of experimental accuracy.

Hole Fermi surface. The value of the effective mass, the behavior of which is represented by curve 4 of Fig. 3, corresponding to a magnetic

Authors	Measurement method	Ratio of axes of the ellipsoid*
Dillon, Shoenberg, 1955 [3]	de Haas–van Alphen effect	10
Brandt, Ventsel', 1958 [11]	The same	(8–8.6) sec 30° = 9.2–9.9
Brandt, Razumeenko, 1960 [11]	»	10–11
Weiner, 1962 [8]	»	7.9 sec 30° = 9.1
Galt et al, 1959 [9]	Cyclotron resonance	14.3
Aubrey, 1961 [12]	The same	13.3
Present investigation	»	14.8

*The ratios of the ellipsoid axes were determined from the experimental plots published in the indicated articles, and are of corresponding accuracy.

field directed along the C_3 axis, practically corresponds with the effective mass of the holes as measured by Galt et al [9] for the same direction. The corresponding data for the direction along the C_2 axis differ by ~20 per cent. This discrepancy, however, cannot be attributed, for example, to errors in the specimen orientation. Thus, it appears justified to ascribe to the holes an effective mass whose anisotropy is represented by the experimental curve 4. A certain difference between the two branches of this curve, lying in the different quadrants in Fig. 3, is essentially the consequence of the inaccuracy in the specimen orientation, corresponding to the right quadrant of Fig. 3.

Judging from curve 4 of Fig. 3, the form of the hole Fermi surface can be close to an ellipsoid of revolution with a major axis parallel to the C_3 axis, with an axis ratio on the order of 3.5 (mass ratio). However, as can be seen from Fig. 3, two other masses are observed in the basal plane, represented by curves 5 and 6 (the latter apparently has curve 7 as its continuation), the isotropic behavior of which coincides with the behavior of the hole mass 4. It is therefore probable that masses 5 and 6 also pertain to the hole Fermi surface. But in this case the simplest model of this surface in the form of an ellipsoid of revolution cannot be conserved: it is necessary to modify it in such a way as to cause the appearance of the singularities responsible for the cyclotron resonances[4)] 5—7 (Fig. 3). In this respect certain possible variants of the form of the hole Fermi surface for bismuth, considered by Abrikosov and Fal'kovskiĭ [1], are of interest.

It must be noted that the ratio of the axes of the hole "ellipsoid," calculated as the ratio of the areas of its cross sections, as obtained by Brandt [10], amounts to ~3.8, which practically coincides with the mass ratio ~3.5 given above. However, in view of the statements made above regarding the form of the hole surface, this agreement is apparently accidental.

RESULTS OF INVESTIGATION OF QUANTUM OSCILLATIONS

The origin of the quantum oscillations of the surface impedance has the same physical cause as the admittance oscillations (the Shubnikov–de Haas effect) and the oscillations of the magnetic susceptibility of a metal (the de Haas–van Alphen effect). According to the quasi classical theory [20], these oscillations should be described by periodic functions of the reciprocal field, as is confirmed by experiment.

The main result that can be obtained from investigations of quantum oscillations of the surface impedance of a metal is the determination of the areas of the extremal sections of the Fermi surface by planes perpendicular to the direction of the magnetic field in each experiment.

The area of the section S is calculated from the experimentally measured period ΔH^{-1} of the quantum oscillations, as a function of the reciprocal field, in accordance with the formula

$$S = eh / c\Delta H^{-1}$$

in analogy with the processing of the experiments on the de Haas–van Alphen effect [4,16,20]. Therefore the main problem of the experiment consists of an accurate measurement of the periods of the quantum oscillations.

The values of the periods and their anisotropy are determined from records of the type shown in Fig. 2. The interval of the fields in which the oscillations are observed, and consequently their number, is relatively small. However, by extrapolating the record of an experiment made at $\angle HC_2 \lesssim 2°$ to the origin ($H^{-1} = 0$) it is possible to establish accurately the numbers of the oscillations, the displacement of which is then traced continuously on the records obtained with gradual increase of the angle $\angle HC_2$. This procedure raises the accuracy of the relative measurements and at the same time the accuracy with which the anisotropy of the oscillations is investigated. We note that this does not improve the absolute accuracy of the measurements, since the initial phase of the oscillations is not known beforehand. In the experiments described the initial phase was equal to zero, within the measurement accuracy, and was assumed to be the same on all the records.

It is clearly seen on Fig. 2 that each curve represents a recording of beats between two oscillations, close in period and in amplitude. The "fadings" of the beats (numbered with Roman numerals) appear to the right, as the field is rotated, and move toward the left ends of the curves. The beat period on the upper curve, which exceeds the entire length of the record, was determined also from the continuously traced number of the "fadings," measuring from the origin ($H^{-1} = 0$). We note that the phase (sign) of the oscillation of a given

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[4)]It is also necessary to take into account the fact that in constructing the diagram of Fig. 3 we used only cyclotron resonances which were most pronounced on the experimental curves and accompanied by clearly visible resonances of order higher than the first (except for curve 5). Therefore the diagram of Fig. 3 can be supplemented in further experiments with some new data.

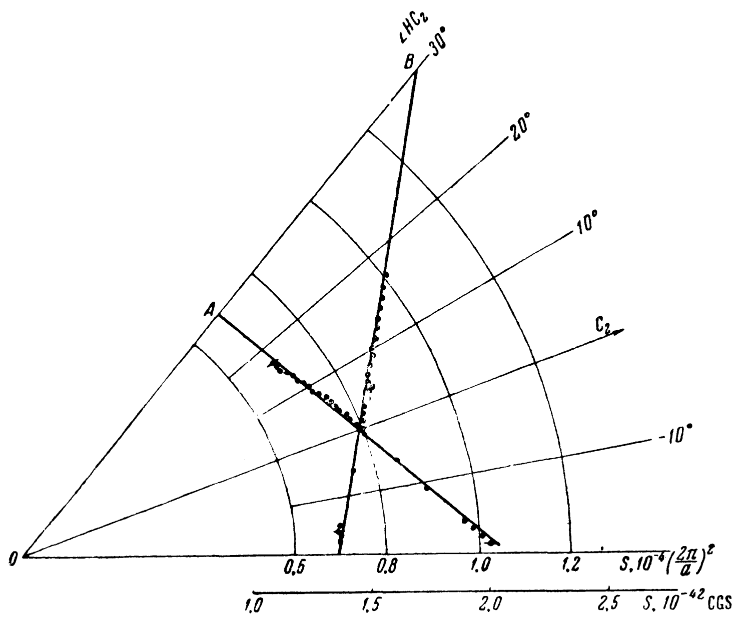

FIG. 5. Areas of extremal sections of electronic Fermi-surface ellipsoids of bismuth, determined from the quantum oscillations for different directions of the magnetic field in the basal plane (the angle is reckoned from the binary axis C_2). a – Lattice constant of bismuth.

number, for example 14 on Fig. 4, changes after the "fading" of the beats passes through its location. The described procedure of measurements makes it possible to determine with high accuracy the areas of the two sections of the Fermi surface responsible for the appearance of two oscillations of close period, and the difference in the areas of these sections is determined from the periods of the beats with the same relative accuracy as the sections themselves (this leads to a clearly pronounced character in the spread of the experimental points, which is seen on Fig. 5 and particularly on Fig. 6, namely each pair of points representing two sections in a given direction deviates to one side of the straight lines).

In the present investigation quantum oscillations of the surface impedance of bismuth were investigated at magnetic field directions close to the direction of one of the binary axes (C_2) of the bismuth. In this case the oscillations were observed only in sections of nearly equal area of two electronic ellipsoids, the major axes of which were perpendicular to the other two axes C_2. On the other hand, the oscillations on the ellipsoid which is prolate perpendicular to the first C_2 axis, were not observed because they should have a period which is one order of magnitude smaller, and occurs at a magnetic field intensity which is one order of magnitude larger, that is, in the region 5–7 kOe. At such fields, observation of these oscillations is extremely difficult, owing to the presence of bismuth surface-impedance oscillations of a different origin; they manifest themselves at a field of 1 kOe (Fig. 2) and reach a relatively large amplitude [17].

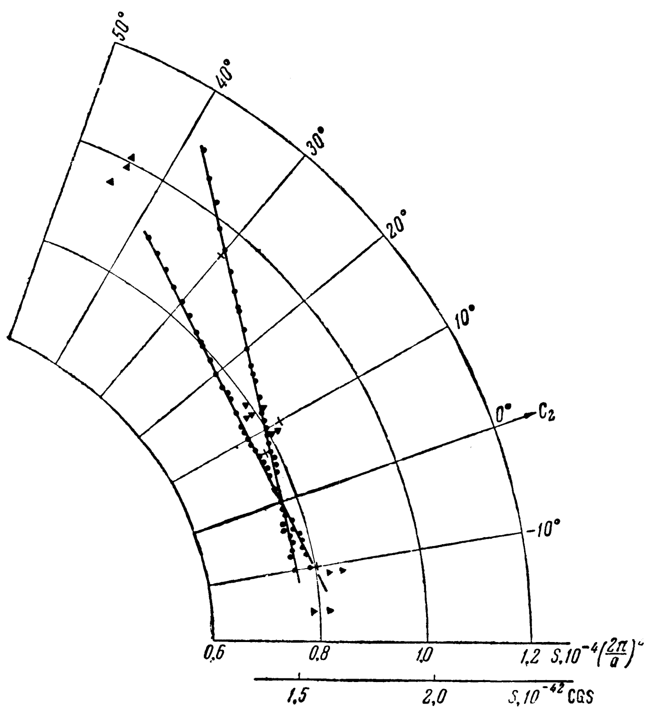

FIG. 6. Areas of extremal sections of electronic ellipsoids of the Fermi surface of bismuth, determined from the quantum oscillations, in a plane containing the binary C_2 and the trigonal axes. The lines were drawn through the experimental points by least squares; the angle between the lines is 14°. The crosses denote the measurement results of Brandt and Ventsel',[11] the triangles the results of Weiner[8] (a – lattice constant of bismuth).

Figure 5 gives the results of the measurement of the extremal areas of intersection between the electronic ellipsoids of the Fermi surface and planes parallel to the trigonal axis of the crystal. This figure must be compared with Fig. 4, which shows the effective masses of the electrons moving in phase space along orbits that are the perimeters of the Fermi-surface sections shown in Fig. 5. Information on the location and form of the electronic ellipsoids, which follows from Fig. 5, coincides with the statements made above on the basis of an analysis of Figs. 3 and 4: the major axes of the three ellipsoids are perpendicular to the binary axes C_2; the central parts of the ellipsoids are indistinguishable in form, within the measurement accuracy, from right cylinders.

The absence in Fig. 5 of experimental points at angles $\angle HC_2 > 19°$ is due to the fact that the observation of the quantum oscillations was practically impossible under these conditions, owing to the rapid growth in the amplitude of other oscillations with smaller period, the appearance of which is noted on Fig. 2. The investigation of the quantum oscillations at angles $\angle HC_2 \leq 30°$ was carried out by Vol'skiĭ[21] (at a frequency ~ 3 Mc), who established that the lines drawn in Fig. 6 represent correctly the sections of the ellipsoids up to $\angle HC_2 = 30°$, that is, up to a field direction along the trace of the symmetry plane σ_d of the bismuth crystal (thus, OB = 2OA in Fig. 6).

From a comparison of Figs. 4 and 5 it follows directly that within the range $\angle HC_2 \leq 30°$ the electron effective mass if proportional to the area of the extremal Fermi-surface section enclosed in the orbit of these electrons in phase space. This is experimental proof of the fact that within the indicated limits the deviation of the dispersion law of the bismuth electrons from quadratic is smaller than the measurement errors (in the analysis of the investigations of cyclotron resonances above it was established that the quadratic dispersion law cannot hold true for the entire electronic Fermi surface). On this basis we can calculate the limiting energy of the bismuth electrons:

$$E_0 = S/2\pi\mu m_e = (2.5 \pm 0.1)\cdot 10^{-14} \text{ erg},$$

which corresponds to an effective temperature 181 ± 7°K. The limiting electron velocity, pertaining to the central part of the ellipsoidal Fermi surface, is

$$v_0 = \sqrt{2E_0/\mu m_e} = (7.7 \pm 0.2)\cdot 10^7 \text{ cm/sec}.$$

The results of the experiments shown in Fig. 6 yield the inclination of the major axes of the electronic ellipsoids to the basal plane of the crystal. The experimental points of Fig. 6 were obtained by an analysis of records (similar to those of Fig. 2) of quantum oscillations at different magnetic-field directions in the plane containing the trigonal and binary axes. The angle between the lines drawn in Fig. 6 is equal to the projection of the angle between the major axes of the ellipsoids on the symmetry plane σ_d of the crystal. The angle of inclination of the major axes of the ellipsoids to the basal plane turns out to be 6° ± 15′.

The results of the investigations of the quantum oscillations of the surface impedance of bismuth are in good agreement with the model proposed by Shoenberg[2,3] for the electronic Fermi surface of bismuth. To evaluate the accuracy attained and the degree of detail in the measurements, and consequently the degree of accuracy and reliability of the quantitative conclusions made in comparison with the available experimental data, Fig. 6 shows the results of Weiner[8] and Brandt and Ventsel'[11]. It is possible to make an analogous comparison on Fig. 5, since this crystallographic plane was not investigated so far by anyone. On the other hand, a comparison of the results of the experiments obtained by measuring the de Haas–van Alphen effect[3,8,11] in the bisector symmetry plane σ_d of the bismuth crystal (which was not investigated in the given work) discloses an even larger scatter of the experimental points and greater discrepancies between data of different authors, than is the case for the plane of Fig. 6. Therefore a comparison of the quantitative results of the present work with the literature data can serve only as a check on the latter.

In constructing his model for the electronic surface, Shoenberg [3] assumed that the angle of inclination of the major axes of the ellipsoids to the basal plane of the bismuth crystal is 5°45′. Further experiments[8,9,12] were in good agreement with the values of the angle of inclination, within a range 5°41′ — 6°10′ (the numbers are cited without an account of the measurement errors). These values practically coincide with the angle measured in the present work, namely 6°00′ ± 15′. The value of the limiting electron energy is in worse agreement: Shoenberg[4] gives a value 2.8×10^{-14} erg, whereas in our work we obtained a limiting energy of $(2.5 \pm 0.1) \times 10^{-14}$ erg.

CONCLUSION

In order to make a detailed quantitative investigation of certain singularities of the Fermi surface of bismuth, we investigated two effects that manifest themselves in changes of the surface impedance of single crystals of pure bismuth at microwave frequencies, namely cyclotron resonance and quantum oscillations. The quantum oscillations of the surface impedance of the metal have not been investigated experimentally heretofore.

An investigation of both effects was made on the same specimens by a common method, essentially in one and the same experiment, which makes the quantitative deductions obtained by comparing the results of the measurements of these two effects quite reliable. The high accuracy and the degree of detail of the data obtained was guaranteed by the high resolution and sensitivity of the measurement method.

Investigations of cyclotron resonance gave detailed information on the anisotropy of the effective masses of the current carriers in the bismuth in two crystallographic planes. An investigation of the quantum oscillations made it possible to study in detail the dependence of the cross section areas of the electronic ellipsoids of the Fermi surface of bismuth on the direction in a region adjacent to the binary axes in the two crystallographic planes. An analysis of the experimental results pertaining to electrons has shown good agreement with the electronic model of the bismuth Fermi surface, proposed by Shoenberg. The limits of the validity, within the precision of the experiment, of the quadratic dispersion law for the electrons were determined and it was established that this law is not satisfied outside these limits. The accuracy and degree of detail of the experimental data greatly exceed the corresponding information existing in the literature.

It must be particularly emphasized that the simultaneous study of two phenomena most sensitive to the geometry of the Fermi surface by using the same specimens is methodologically highly convenient. Although this is an obvious consideration, no such experiments have been carried out until now.

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