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INVESTIGATION OF THE COEFFICIENT OF ABSORPTION OF SOUND IN BISMUTH. II. GIANT OSCILLATIONS

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The giant oscillations of the coefficient of absorption of longitudinal ultrasound in single-crystal samples of bismuth were investigated at 1.4°K. The measurements of the oscillation periods were carried out at the frequencies of 220 and 300 Mc along three principal crystallographic planes. The profiles and amplitudes of the absorption lines are in good agreement with the theoretical predictions. The experimental results obtained were compared with the results of investigations by other methods.

THE interaction of the acoustical phonons with the conduction electrons in metals appears most clearly at low temperatures, when the mean free path of electrons l becomes longer than the phonon wavelength λ. In a magnetic field, the absorption coefficient of sound Γ exhibits oscillations whose nature depends on the range of fields applied and on temperature. In relatively weak fields, for which $\hbar\Omega < kT$ ($2\pi\hbar$ is Planck's constant, Ω is the cyclotron frequency, k is Boltzmann's constant, T is the temperature), the geometrical resonance is observed whose measured period can be used to determine the extremal diameter of the Fermi surface. This resonance has already been investigated in bismuth by the present author.[1] In stronger magnetic fields $\zeta < \hbar\Omega < kT$ (ζ is the chemical potential of the electron gas), quantum oscillations of the absorption coefficient of sound are observed and their amplitude becomes "giant"—considerably higher than the value of the absorption coefficient Γ_0 in the absence of a magnetic field—when the vectors K and H are not perpendicular (K is the wave vector of the sound, H is the magnetic field vector).

The giant oscillation effect was predicted theoretically by Gurevich, Skobov, and Firsov.[2] The effect has been detected experimentally in zinc,[3] bismuth,[4,5] and gallium.[6,7] The theory has been developed further in other papers.[8-11]

According to the theory, the oscillation period ΔH^{-1} should be constant when considered as a function of the reciprocal field and should be governed by the characteristic parameters of the Fermi surface:

$$\Delta H^{-1} = 2\pi e\hbar / cS(\zeta, p_{H0}), \tag{1}$$

where the quantity p_{H0} is related to the relative positions of the vectors K and H:

$$p_{H0} = m_H s|\mathbf{K}||\mathbf{H}| / (\mathbf{KH}). \tag{2}$$

In these formulas, c is the velocity of light; s is the velocity of sound; S is the area of the section of the Fermi surface cut by the plane p_{H0}, normal to the vector H; e is the electron charge; m_H is the "longitudinal" electron mass.

Thus, in principle, by varying the orientation of the vectors K and H, we can measure the area of any section of the Fermi surface and not just the extremal sections, as is the case in the de Haas—

van Alphen effect. However, in practice, it is difficult to make use of this property since, because of the low value of the velocity of sound, the quantity p_{H0} remains close to zero over a considerable range of the values of the angle ϑ between **K** and **H**.

This does not reduce the interest in the investigation of the giant oscillations. Because of the large amplitude, the method is very sensitive, and in some cases we can obtain additional information about the electron spectrum of the investigated metal by measuring directly the values of the electron—phonon coupling constant Λ_{ik}.

The present investigation is a continuation of studies of the electron spectrum of bismuth. It seemed of interest to compare the experimental data obtained by the geometrical resonance method and those found from the giant oscillations, as well as to investigate the features of the anisotropy of the absorption of sound in various crystallographic planes.

MEASUREMENT METHOD

We used a pulse method[12] to measure the periods and amplitudes of the giant oscillations. The high-frequency sound was excited by generating harmonics in piezoelectric X-cut quartz plates whose resonance frequency was 20 Mc. The main results were obtained at the frequency of 220 Mc, but some measurements were carried out under continuous-operation conditions at the frequency ν = 300 Mc employing magnetic field modulation and synchronous detection.

A cryostat, in which the inclination of the magnetic field with respect to the wave vector direction could be varied, is shown in Fig. 1.

The samples were cut by electric-spark machining from a large single crystal grown from the melt in a Pyrex ampoule; they were in the form of cubes, the normals to the cube faces being parallel to the crystal axes. The dimensions of a sample were 5 × 5 × 5 mm. Fine abrasives on a grinding plate were used to make the sample faces strictly plane parallel. The method of locating a sample, stuck to a special brass piston ring, can be seen from Fig. 1. The lower part of the crystal holder had special apertures for the exact alignment of the crystal, with respect to the axis of the instrument, by means of an optical goniometer.

In this instrument, the wave vector of the sound lay in a horizontal plane, which included the vector of the constant field **H** established by means of an electromagnet on a rotating base. Much attention was paid to reducing the dimensions of the working part of the crystal holder in order to obtain the

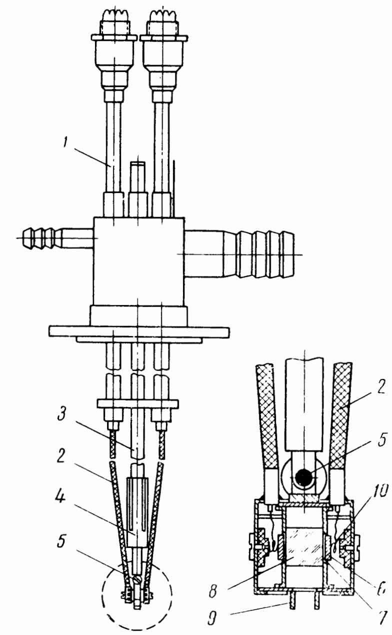

FIG. 1. Cryostat and crystal holder for the investigation of the absorption of sound in an inclined magnetic field: 1) coaxial lines; 2) flexible coaxial couplings; 3) guide rod; 4) split spring coupling; 6) electrode; 7) piezoelectric quartz transducer; 8) sample; 9) aperture for alignment of a crystal by means of an optical goniometer; 10) spring contacts.

highest possible magnetic field intensities. The internal diameter of the nozzle of the helium Dewar flask used was 15.5 mm, which made it possible to obtain a maximum magnetic field intensity of 12 500 Oe.

The oscillations, recorded as a function of the reciprocal of the magnetic field, were subjected to the following analysis. The measurement of the periods of the giant oscillations was made easier by the fact that the absorption lines, particularly in strong magnetic fields, were much narrower than the separations between the lines. Instead of an analysis of the beats, used in such cases, it was frequently sufficient to measure simply the appropriate separations between the absorption peaks in the reciprocal field.

The measurements were carried out on two samples: a main sample and a control sample. Since the results of the control measurements were practically identical with those obtained for the main sample, only the results of the measurements for one sample are given in the present paper.

EXPERIMENTAL RESULTS

1. Oscillation amplitude. For convenience, we shall introduce a system of rectangular coordinates xyz, with x along a binary axis, y along a bisector axis, and z along a trigonal axis of a crystal.

Figure 2 shows a recording of the spectrum of

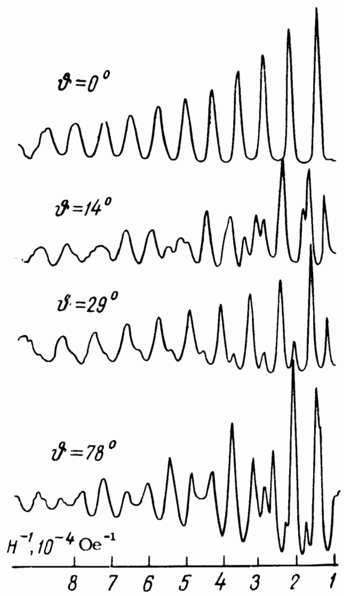

FIG. 2. Giant oscillations in the binary axes plane, recorded as a function of the reciprocal magnetic field. The ordinate gives the absorption coefficient in arbitrary units. ϑ is the angle between the wave vector of sound **K** and the magnetic field vector **H**; T = 1.4°K; ν = 220 Mc; **K** vector parallel to the binary axis.

the giant oscillations for sound propagated along the x axis with the magnetic field lying in the xy plane (ϑ is the angle between K and H; in this case, it was the angle between the x axis and the vector H).

A characteristic feature of the giant oscillations was the presence of narrow absorption lines against a background of wide diffuse minima, as well as a strong angular dependence of the amplitude near $\vartheta \approx 90°$. Figure 3 shows the angular dependence of the amplitude (in arbitrary units) for three orientations of the vector K. A minimum was observed at $\vartheta = 90°$ but the amplitude did not decrease to zero in all cases. The same behavior of the oscillations was reported in[5]. 1)

Figure 4 shows the dependence of the oscillation amplitude on the magnetic field when the vectors K and H were parallel to the x axis. According to the theory.[2,9] the oscillation amplitude should depend linearly on the magnetic field

$$\Gamma_m = \Gamma_0^{(i)} e\hbar H / 8m^* ckT. \qquad (3)$$

1)The earlier paper [1] gave figures with recordings of the geometrical and quantum oscillations of the absorption coefficient of sound in bismuth for **K** ⊥ **H** (Fig. 3). The high amplitude of the quantum oscillations was due to the high sensitivity of the apparatus under continuous-radiation conditions, with field modulation and synchronous detection, and also possibly due to small accidental deviations of the vectors **K** and **H** from normal orientation with respect to each other (for the geometrical resonance, such deviations are unimportant).

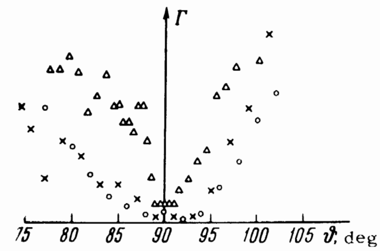

FIG. 3. Angular dependence of the amplitude of oscillations near $\vartheta \approx 90°$ (ϑ is the angle between **K** and **H**). The ordinate gives the amplitude in arbitrary units; T = 1.4°K; ν = 220 Mc. △ – **K** ∥ x, **H** close to the y axis; ○ – **K** ∥ y, **H** close to the x axis; × – **K** ∥ y, **H** close to the z axis.

In this formula, $\Gamma_0^{(i)}$ is the electronic absorption coefficient in the absence of a field, representing that group of carriers which contributes to the oscillations (no allowance for the spin degeneracy).

The formula (3) allows us to estimate the value of $\Gamma_0^{(i)}$ using for this purpose the slope of $d\Gamma/dH$ of the experimental straight line in Fig. 4. Two electron ellipsoids contributed to the oscillatory curve for H ∥ x and, consequently, using the known values of the effective mass $m^* = 0.01m_0$, we obtained the value of $\Gamma_0^{(1)}$ at the longitudinal sound frequency $\nu = 220$ Mc: $\Gamma_0^{(1)} = 0.135$ cm^{-1} per ellipsoid.

To determine the local value of the electron–phonon interaction constant, we can use the formula (28) from[9]:

$$\Gamma_0 = \frac{m^2 |\Lambda_{ik} u_{ik}|^2}{2\pi\rho s\hbar^3 |\mathbf{u}|^2 |\mathbf{K}|}, \qquad (4)$$

where ρ is the density of the crystal, u_{ik} is the deformation tensor, $m^2 = m^* m_H$ is the product of the cyclotron mass and the "longitudinal" mass $(1/m_H = \partial^2\epsilon/\partial p_H)$, **u** is the displacement vector of the acoustic wave. After carrying out the necessary calculations, we found $|\Lambda_{xx}| = 3.31$ eV per ellipsoid.

2. Oscillation Period. The Shoenberg–Brandt model of the electron spectrum of bismuth, usually employed in the calculations, consists of three

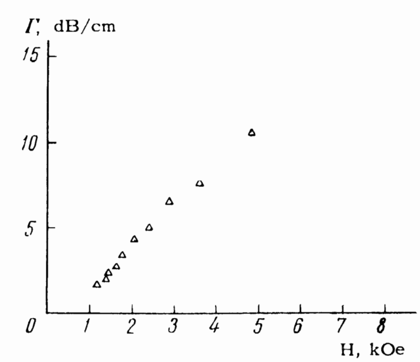

FIG. 4. Dependence of the oscillation amplitude on the magnetic field for **K** ∥ **H** ∥ x, ν = 220 Mc, T = 1.4°K.

electron ellipsoids lying symmetrically with respect to a trigonal axis. The period of the giant oscillations can be found easily within the framework of this model.

We shall now write the area of the section of an ellipsoid by a plane p_n, normal to the unit vector $\mathbf{n} = \mathbf{H}/|\mathbf{H}|$:

$$S(\varepsilon_f, p_u) = 2\pi m_0\varepsilon_f\left(\frac{\det m}{\mathbf{n}m\mathbf{n}}\right)^{1/2}\left[1-\frac{p_n^2}{2m_0\varepsilon_f(\mathbf{n}m\mathbf{n})}\right] \quad (5)$$

where m is the mass tensor. The value of p_n at which the oscillations are observed is found by assuming that the projection, onto the wave vector **K**, of the average electron-drift velocity along the field is equal to the velocity of sound s:

$$v_{\mathbf{K}} = \frac{\cos\vartheta}{2\pi m^*}\frac{\partial S(\varepsilon_f, p_n)}{\partial p_n} = s. \quad (6)$$

Combining the obtained value with Eqs. (5) and (1), we find

$$\Delta H^{-1} = \frac{e\hbar}{m_0 c\varepsilon_f}\left(\frac{\mathbf{n}m\mathbf{n}}{\det m}\right)^{1/2}\left[1-\frac{m_0 s^2(\mathbf{n}m\mathbf{n})}{2\varepsilon_f\cos^2\vartheta}\right]^{-1}. \quad (7)$$

The results of the measurements of the oscillation periods along the investigated directions of the vector **K** are given below.

A. The vector **K** parallel to the x axis. Figure 5 shows the stereographic projection of the planes (dashed curves) in which the magnetic field vector was rotated and in which the giant oscillations were investigated. A detailed analysis of the oscillograms was carried out only for the principal crystallographic planes. For the planes inclined at 6, 30, and 60° to the basal plane, we determined the positions of the points of coincidence of the oscillation periods associated with different ellipsoids. The positions of the lines with equal periods, which could be found from Eq. (7), depended only on the

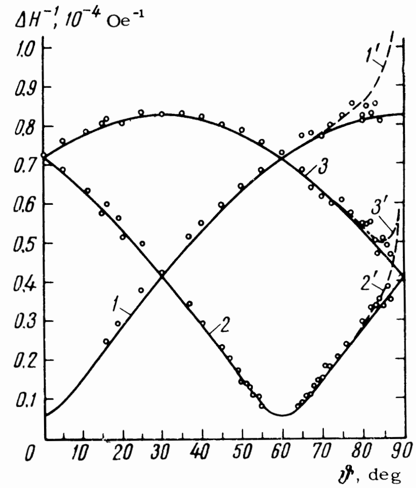

FIG. 6. Angular dependence of the oscillation periods in the binary axes plane. Continuous curves 1, 2, 3 give calculated values for the central cross sections; dashed curves 1', 2', and 3' were obtained from Eq. (7) allowing for the difference between the measured and central cross sections. The parameters used in the calculation were taken from [1]. **K** ∥ x, angle measured from the x axis.

angle of inclination ξ of the principal axis of an ellipsoid to the basal plane:

$$\sin(\varphi - 60°) = \tan 2\xi / \tan\theta. \quad (8)$$

Here, φ and θ are the spherical coordinates of the vector **n**. The thick curves in Fig. 5 were plotted using Eq. (8) and assuming that $\xi = 6°$. The experimental points coincided quite well with these curves. Examples of oscillations in the binary axes plane are given in Fig. 2. A characteristic feature of the oscillograms for **K** ∥ x was the presence of components of three electron ellipsoids.

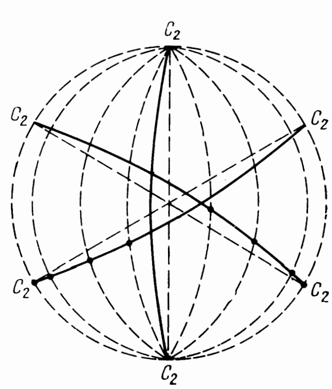

FIG. 5. Stereographic projection of equal-period lines (continuous curves); C_2 points of emergence of the binary axes. Dashed curves indicate projections of the planes in which the positions of the points of coincidence of the periods were measured for **K** ∥ C_2.

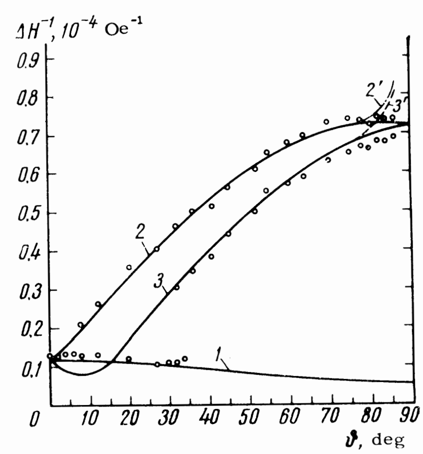

FIG. 7. Angular dependence of the oscillation periods in the plane of the trigonal and binary axes. Continuous curves 1, 2, 3 were calculated for the central cross sections; dashed curves 2' and 3' were plotted using Eq. (7) and allowing for the difference between the measured and central cross sections. **K** ∥ z, angle measured from the z axis.

An analysis of the periods of the beats in the xy plane is given in Fig. 6.

Using the parameters of the spectrum given in[1] and Eq. (7), we can calculate the periods of the giant oscillations as a function of the magnetic field direction. Curves 1—1′, 2—2′, and 3—3′ give the results of the calculation for the binary axes plane. The experimental values of the periods are in good agreement with the calculated values in the angular range $0° < \vartheta < 80°$. For $\vartheta > 80°$, there is a noticeable divergence from the calculated values.

B. The vector **K** parallel to the z axis. The results of the measurements of the oscillation periods in the zx plane are given in Fig. 7. For the longitudinal sound propagated along the z axis, there is again an interaction with three electron ellipsoids, but the amplitude of the oscillations depends strongly on ϑ: the ellipsoids 1, in the range $35° < \vartheta < 90°$, and 2, in the range $0° < \vartheta < 35°$ make negligibly small contributions.

Curves 1, 2—2′, 3—3′ in Fig. 7 were plotted using Eq. (7) and the parameters given in[1]. In the angular range $\vartheta > 80°$ in the zx plane, there is again a considerable divergence between the calculated and measured values of the oscillation periods.

C. The vector **K** parallel to the y axis. Examples of recordings of the giant oscillation spectrum for $\mathbf{K} \parallel y$ in the binary-axes plane are given in Fig. 8. The interaction between the longitudinal sound wave having its wave vector parallel to the bisector axis and electrons has an interesting feature:

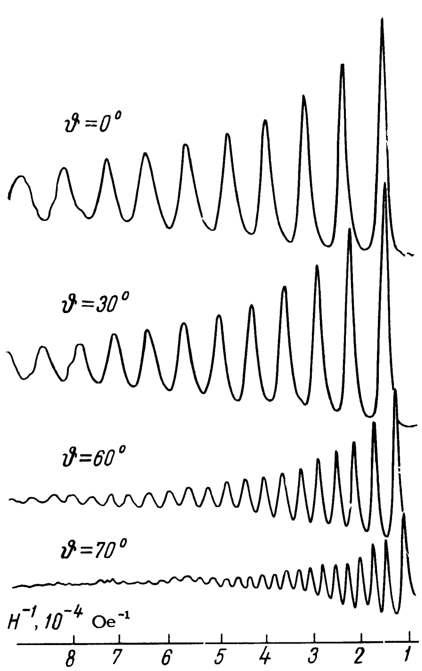

FIG. 8. Giant oscillations in the binary axes plane for $\mathbf{K} \parallel y$. $T = 1.4°K$, ϑ is the angle between **K** and **H**, $\nu = 220$ Mc.

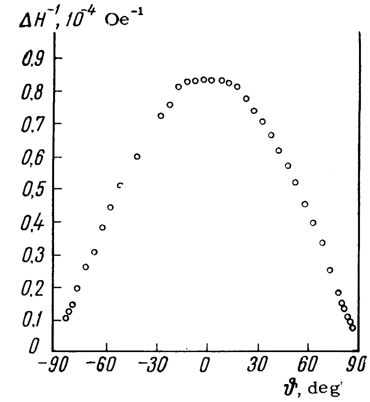

FIG. 9. Angular dependence of the oscillation period in the binary axes plane for $\mathbf{K} \parallel y$. The angle is measured from the y axis.

its magnitude is considerably greater for that electron ellipsoid which is elongated along the axis of propagation of the sound wave. The sound hardly "notices" other carriers and the oscillation period is governed only by this ellipsoid for any direction of the magnetic field.

The results of the measurements of the periods in the xy and yz planes are given in Figs. 9 and 10. For any orientation of **H**, there was, in practice, only one oscillating component, with the exception of the yz plane for $|\vartheta| > 60°$, for which there was one more oscillation period, evidently associated with holes.

In the binary axes plane, the periods coincided, within the limits of the error, irrespective of whether the vector **K** is directed along the x axis or the y axis. In the yz plane, the electron oscillations in the range of large angles were of greatest interest. In this range of angles, the de Haas—van Alphen oscillations disappeared even at very low temperatures.[13] The amplitude of the giant os-

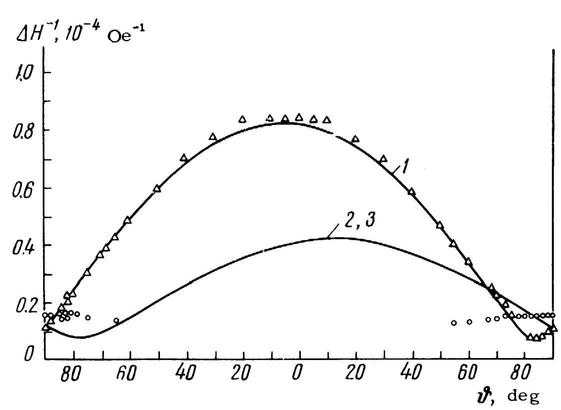

FIG. 10. Angular dependence of the oscillation period in the yz plane for $\mathbf{K} \parallel y$. Continuous curves 1, 2, 3 give the calculated values of the periods for the central cross sections of the electron ellipsoids. The angle was measured from the y axis. △ — electron oscillations; ○ — oscillations evidently associated with the hole surface.

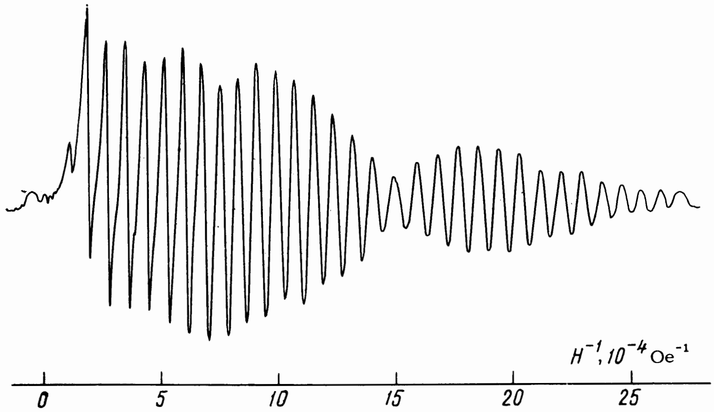

FIG. 11. Recording of the derivative of the absorption coefficient as a function of the reciprocal magnetic field for $\mathbf{K} \parallel y$ and $\vartheta \approx 6°$ in the yz plane. T = 1.4°K, ν = 300 Mc.

cillations in this region was also much smaller and the oscillations disappeared in the range $76° < \vartheta < 82°$. The minimum of the period was evidently reached near $\vartheta = 84°$ and this made it possible to measure the area of the "average" cross section of the electron ellipsoid. The measurements in the yz axes plane were carried out using a continuously operating spectrometer at the sonic frequency ν = 300 Mc. Under these conditions, the sensitivity of the apparatus was considerably higher.

The theory[14,15] admits the possibility of the existence of a "constriction" in the electron surface detectable from the beats of the periods for H near the y axis (near ΔH^{-1}_{max} in the yz plane).

Using the maximum sensitivity of the apparatus, we were able to obtain about 35 absorption lines (Fig. 11) for this direction of the magnetic field. In a field of about 700 Oe, we indeed observed a beat "waist" but a detailed investigation showed that the position of the "waist" shifted with the frequency of the sound. The nature of this effect was, therefore, not associated with the shape of the Fermi surface.

DISCUSSION OF RESULTS

Gurevich, Skobov, and Firsov [2,9] obtained the condition for the existence of giant oscillations: $(Kl)^2 \gg \zeta/\hbar\Omega$. This relationship was satisfied by the investigated bismuth samples at temperatures $T \approx 1.5°K$ and sonic frequency $\nu \approx 200$ Mc, in magnetic fields H > 20 Oe. The oscillation amplitudes and the absorption line profiles were in sufficiently good agreement with the experimental data. The present author [4] has carried out a comparison of the calculated and experimentally recorded oscillation curves for bismuth. In spite of the fact that the calculation was carried out for a spherical model of the Fermi surface and the bismuth spectrum was in fact far removed from such an approximation, the calculated and experimental curves were practically identical: the nature of the energy spectrum did not greatly affect the line profile.

A strong dependence of the oscillation amplitude on the angle was found experimentally near $\vartheta \approx 90°$. The quantum oscillations for $\vartheta = 90°$ (i.e., $\mathbf{K} \perp \mathbf{H}$) were of low amplitude. Under such conditions, electrons can absorb sound only during their collisions with scatterers and the oscillation amplitude should be $1/Kl$ times smaller than the giant amplitude. A non-zero oscillation amplitude for $\mathbf{K} \perp \mathbf{H}$ was also reported by Toxen and Tansal[5] but, due to the smaller value of Kl (the frequency of sound ν used by them was 40 Mc[5]), their ratio of the amplitudes was, as expected, smaller. However, the problem of the quantitative relationship between $\Gamma(H_\perp)$ and $\Gamma(H_\parallel)$ still requires more rigorous theoretical discussion.

The value obtained for the electron—phonon interaction constant $|\Lambda_{xx}|$ agreed, within the limits of the experimental error, with the values of the deformation potential $E_1 = -2.4$ eV and $E_2 = +2.5$ eV, measured by the static piezoresistance method.[16] The purpose of the present study was not to determine the total vector $|\Lambda_{ik}|$. We can point out here only that the method of measuring the electron-phonon interaction constants with the help of the giant oscillations is very promising because it is known immediately with which group of carriers the values obtained are associated (unfortunately, the sign of Λ_{ik} cannot be determined).

Thus, bearing in mind the results of the present investigation, we may assume that the amplitude characteristics of the giant oscillations are in sufficiently good agreement with the theoretical predictions, but a quantitative comparison with the theory would require an accurate determination of Γ_0. Toxen and Tansal[5] obtained anomalous values

$S_1 \cdot 10^{42}$	$S_2 \cdot 10^{42}$	$S_3 \cdot 10^{42}$	ξ, deg	Reference	Remarks
1.31 ± 0.1	13.1 ± 1.0	18 ± 1.5	6	[1]	Ellipsoidal model calculations
1.28 ± 0.065	13.3 ± 0.65	19 ± 2	6	Present study	S_3 extrapolated
1.34 ± 0.05	14 ± 1.5	19.5 ± 1		[13,18]	S_2 extrapolated
1.19			4	[19]	

of the ratio Γ/Γ_0 because of an inaccurate estimate of Γ_0.

It is very difficult to measure independently Γ_0 of a metal which does not go over to the superconducting state, because the experiment always gives the sum $\Gamma = \Sigma\Gamma_{0i} + \Sigma\Gamma_l$ of the electronic and lattice absorption coefficients.

The oscillation periods near the angles $80° < \vartheta < 90°$ in the xy and zx planes differed considerably from those calculated in accordance with the quadratic model of the spectrum. The divergences were not large but they were greater than the experimental error. Since the oscillations in this angular range remained of the giant type (we can, in general, assume that for some reason their nature may change and they may go over to the de Haas—van Alphen type, i.e., they may become oscillations of the central cross section), then, because the experiments carried out indicated the absence of a "saddle point" or a "constriction" of the electron surface (the areas S_{max} and S_{min} differ, in the extreme case, by less than 1/30), it was necessary to assume the existence of a longer cylindrical part than was the case for an ellipsoid.

This assumption was in qualitative agreement with the conclusions of Édel'man and Khaĭkin[17], deduced from an investigation of the cyclotron resonance at limiting points although deviations from the ellipsoidal shape were very small. We noted that the cyclotron masses were much more "sensitive" to the nature of the electron spectrum $\epsilon(p)$ than to the shape of the constant-energy surface and, therefore, the difference between the "extremal" and "limiting" points amounting to 35% in[17] was fully compatible with the very slight departure from the ellipsoidal shape.

By way of illustration, the table below gives the results of a comparison of the measured values of the areas of the principal ellipsoid cross sections S_1, S_2, and S_3 (the dimensions of these areas are $g^2 \cdot cm^2 \cdot sec^{-1}$) with those calculated in[1] from the ellipsoidal model.

The angle of inclination of the ellipsoid axis to the basal plane was close to 6° and this value was in good agreement with the results for an arbitrary direction of the vector **H** with respect to the crystal axes and not only with the results for the principal planes (cf. Fig. 5). The largest difference between the two sets of data given in the table was found for the cross section S_3 although even in this case the divergence was within the limits of the experimental error.

The samples used in the investigations of the giant and geometrical oscillations were cut from one large single crystal and, therefore, they had the same amounts of impurities. The good agreement between the results of the investigations by these two methods was, to a considerable degree, due to this factor. The different values of the areas of the cross sections of the Fermi surface of bismuth, reported in a number of papers,[13,19] were probably due to a change in the Fermi level under the influence of impurities.

The oscillations associated with the hole surface were observed in a limited range of angles and this was most likely due to the relatively low value of the ratio H/T reached in our experiments. The measured value of the minimum area of the hole-surface cross section was $S_{1d} = (6.6 \pm 0.3) \times 10^{-42}\ g^2 \cdot cm^2 \cdot sec^{-2}$. The value calculated in[1] was $S_{1d} = (6.0 \pm 0.5) \times 10^{-42}\ g^2 \cdot cm^2 \cdot sec^{-2}$. These values were equal within the limits of the experimental error.

The spin splitting of the Landau levels was not observed in the present study. Investigations of this splitting, using stronger magnetic fields, would undoubtedly be of interest because of the selective sensitivity of sound to different groups of carriers.

In conclusion, the author expresses his gratitude to É. A. Kaner, V. G. Skobov, and V. L. Gurevich, who took part in the discussion of this investigation, and to L. Ya. Matsakov for his help in the measurements.

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