

*SHUBNIKOV-DE HAAS EFFECT IN BISMUTH UNDER A PRESSURE OF 15 KBAR*

E. S. ITSKEVICH and L. M. FISHER

High Pressure Physics Institute, Academy of Sciences, U.S.S.R.

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The influence of uniform compression on the frequency of the quantum oscillations of the electrical resistance (Shubnikov-de Haas effect) of bismuth was investigated at pressures of 11 and 15 kbar, at 1.5°K. A modulation measurement method was used. The angular dependences of the extremal cross sections of the electron and hole ellipsoids were measured at these pressures. The results obtained indicate that the volume of both ellipsoids decreases, under a pressure of 15 kbar, to about one-fifth of its initial value. The hole ellipsoid tends to become spherical under pressure. It is concluded that it would be desirable to observe quantum oscillations, associated solely with the hole part of the Fermi surface, at pressures above 15 kbar.

**S**TUDIES of the influence of uniform compression on the Fermi surface of bismuth at pressures up to 8 kbar have been reported in<sup>[1,2]</sup>. Using a modulation measurement method,<sup>[2]</sup> quantum oscillations of the electrical resistance, associated both with the electron and hole parts of the Fermi surface, have been observed for various directions of the magnetic field with respect to the crystallographic axes of a sample. It has been found that the relative contraction of the small extremal cross sections found for the electron ellipsoid is greater than in the case of the small cross sections of the hole ellipsoid at all the pressures employed. Thus, for example, at a pressure of 7.5 kbar the relative change in the small extremal cross sections of the electron ellipsoid is -42%, but for the hole ellipsoid it is -34%. From these measurements, it has been concluded that the deformation of either the hole or the electron ellipsoid is somewhat anisotropic in the large cross section range.

The present paper reports a continuation of the measurements of quantum oscillations of the electrical resistance of bismuth at pressures up to 15 kbar, carried out to determine the nature of further changes in the volume enclosed by the Fermi surface of bismuth under pressure, as well as the anisotropy of these changes. Direct measurements of the extremal cross sections of the Fermi surface of bismuth at high pressures are of great interest in connection with the postulated electron transition in bismuth<sup>[1-3]</sup> at a pressure of 20-25 kbar.

**METHODS AND SAMPLES**

We used a high-pressure chamber, described in<sup>[4]</sup>, in which pressures up to 18 kbar could be obtained at liquid helium temperatures. The modulation method for measuring the Shubnikov-de Haas effect in a high-pressure chamber is described in<sup>[2]</sup>. In all the experiments, the constant measuring current through a sample was 10 mA and the temperature 1.5°K. The measurements were carried out in a SP-47 electromagnet in magnetic fields up to 14 kOe. The magnetic field was measured with a Hall probe, which was calibrated using a nuclear magnetometer.

The sample used in the measurements was in the form of a slab, 16 × 2.5 × 1 mm, which was cut by the electric erosion method from a single crystal [ $\rho(300^\circ\text{K})/\rho(4.2^\circ\text{K}) \approx 100$ ]. The long axis of the sample coincided with the direction of the bisector axis  $C_1$ . Current and potential contacts were soldered with a nonsuperconducting metal and the sample was placed in a flat triply-wound coil fixed to the valve of the high-pressure chamber. The coil was part of the oscillatory circuit of a Pound-Knight oscillator. This made it possible to measure the quantum oscillations of the electrical resistance also by using a contactless method, at a frequency of about 1 Mc.<sup>[5]</sup> The high-pressure chamber containing the sample was placed in a cryostat, which was located in the electromagnet gap. The magnetic field direction could be varied in the bisector plane of the sample ( $C_2, C_3$ ) by rotating the cryostat.

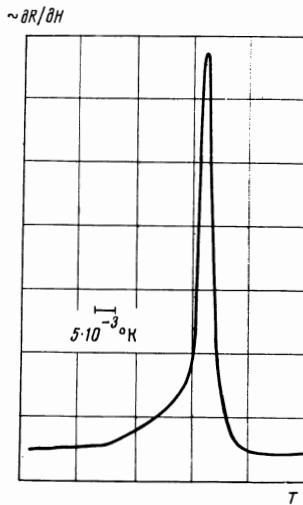


FIG. 1.

The pressure in the chamber was determined from the change in the superconducting transition temperature of tin by a method in which no current is used. For this purpose, we placed a lump of pure tin together with our sample in the coil of the Pound-Knight oscillator and, by slowly pumping a helium bath, we determined the superconducting transition temperature of tin from the sudden increase in the derivative  $\partial R/\partial H$  at the transition point (Fig. 1) ( $R$  is the real component of the surface impedance). At a modulation amplitude of 0.5 Oe ( $f = 23$  cps), the width of the superconducting

transition line did not exceed  $5 \times 10^{-3}$  °K and the error in the value of the pressure was mainly due to the error introduced by the interpolation formula taken from<sup>[6]</sup>:

$$T_{cr}(P) = 3.732 - 4.95 \cdot 10^{-5}P + 3.9 \cdot 10^{-10}P^2.$$

## RESULTS OF MEASUREMENTS

We investigated the dependence of the frequency of the quantum oscillations of the electrical resistance on the angle of rotation of the magnetic field in the  $(C_2, C_3)$  plane ( $\theta = 0$  for  $\mathbf{H} \parallel C_3$ ) at various pressures. The directions of the magnetic field parallel to the binary and trigonal axes were determined from the angular dependence  $[\partial\rho(\theta)/\partial H]_{H=\text{const}}$  with an accuracy of 1–3°. Examples of the recorded electrical resistance oscillations are given in Figs. 2 and 3.

Using magnetic fields stronger than those in<sup>[2]</sup>, we were able to observe oscillations associated with the hole part of the Fermi surface for all directions of the magnetic field in the  $(C_2, C_3)$  plane. For angles  $\theta$  close to 90°, the hole oscillations were observed in magnetic fields exceeding 8–10 kOe and the amplitude of these oscillations decreased, as the pressure was increased, more rapidly than was usually the case (Fig. 2). The frequency of the oscillations and the area of the small cross sections of the electron ellipsoid, propor-

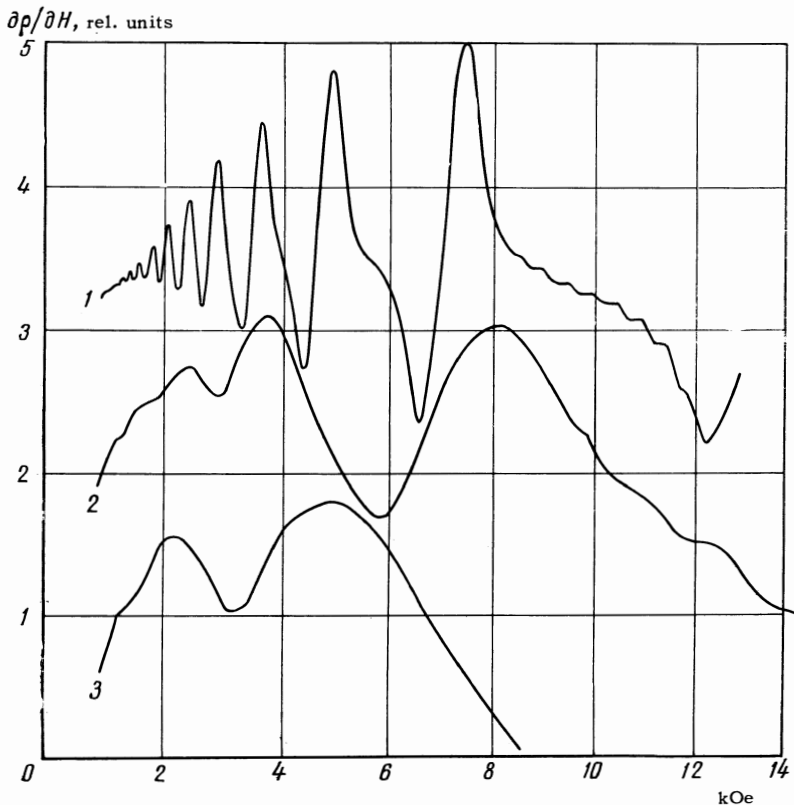


FIG. 2. Examples of the electrical resistance oscillations  $\partial\rho/\partial H$  for  $\mathbf{H} \parallel C_2$  recorded at various pressures. Curve 1 –  $P = 1$  bar, 2 –  $P = 11$  kbar, 3 –  $P = 15$  kbar. Oscillations associated with the small cross section of the electron ellipsoid were observed in  $H < 8$  kOe; curves 1 and 2 for  $H > 8$  kOe show oscillations associated with the large cross section of the hole ellipsoid.

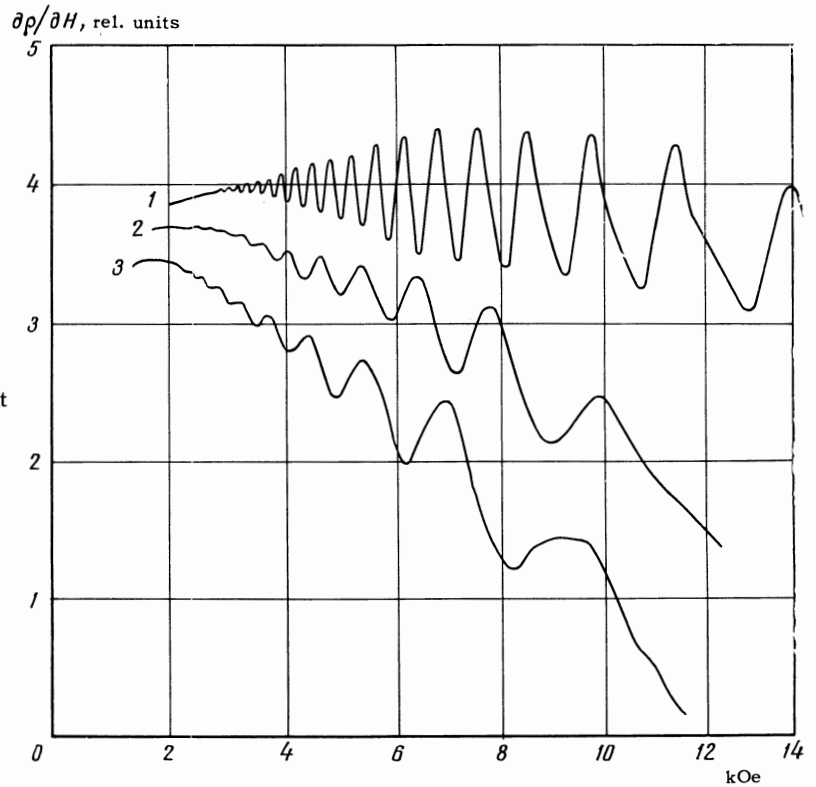


FIG. 3. Examples of the electrical resistance oscillations  $\partial\rho/\partial H$  for  $\mathbf{H} \parallel C_3$ , associated with the small cross section of the hole ellipsoid, recorded at various pressures. Curve 1 –  $P = 1$  bar, 2 –  $P = 11$  kbar, 3 –  $P = 15$  kbar.

tional to this frequency, decreased considerably under pressure. Thus, for example, for  $\mathbf{H} \parallel C_2$  the frequencies were  $\omega_0 = (1.38 \pm 0.03) \times 10^4$  Oe,  $\omega_{15 \text{ kbar}} = (0.41 \pm 0.05) \times 10^4$  Oe. The increase in the oscillation period under pressure complicated the determination of the period and the frequency of oscillations for the small electron cross sections, since over the whole range of magnetic fields we found only three maxima and two minima of  $\partial\rho/\partial H$  at  $P = 15$  kbar (Fig. 2). The maximum error in the determination of the oscillation frequency was 10–12% at all pressures.

The angular dependence of the oscillation frequency  $\omega(\theta)$  is shown, for various pressures, in Fig. 4a for the hole ellipsoid, and in Fig. 4b for the electron ellipsoid. The measured dependence  $\omega(\theta)$  at  $P = 1$  bar is in agreement with the results reported in<sup>[7]</sup>.

We also observed quantum oscillations of the resistance at a frequency of 1 Mc at  $P = 1$  bar. The results of the measurements of the oscillation frequency at high and low frequencies were identical within the limits of the experimental error.

## DISCUSSION OF RESULTS

The relative change in the extremal cross sections of the hole and electron ellipsoids is shown in Fig. 5 for various pressures. As before,<sup>[2]</sup> the

relative decrease under pressure of the small extremal cross sections of the electron ellipsoid were slightly higher than for the small cross sections of the hole ellipsoid: for electrons  $\Delta S/S_0 = 53 \pm 4\%$ , while for holes  $\Delta S/S_0 = 48 \pm 2\%$  at  $P = 11$  kbar, which was the maximum pressure at which all the hole ellipsoid cross sections were measured.

However, it was found experimentally that the largest cross sections of the hole ellipsoid ( $\theta$  close to  $90^\circ$ ) changed under pressure more strongly than the small cross sections (Fig. 5). Hence, we could conclude that the hole ellipsoid tended to become spherical and its volume decreased more.

Our data for atmospheric pressure and for 11 kbar can be used to determine the ratio of the semiaxes  $b/a$  of the hole ellipsoid. At  $P = 1$  bar, as in<sup>[7]</sup>, this ratio is  $b/a = 3 \pm 0.15$ , while under a pressure of 11 kbar, we find that  $b/a = 2.6 \pm 0.3$ . Comparing the results of the measurements of the angular dependence  $\omega(\theta)$  with the results of calculations using the formula

$$\omega(\theta) = \frac{\omega_{min}}{\sqrt{1 + (a/b)^2 \operatorname{tg}^2 \theta \cos \theta}}$$

which is valid for an ellipsoid of revolution, we find that  $b/a = 2.5 \pm 0.3$  at  $P = 15$  kbar. The relative change in  $b/a$  at 11 and 15 kbar is  $(0.13 \pm 0.15) \times 100\%$  and  $(0.17 \pm 0.15) \times 100\%$ , respectively. This

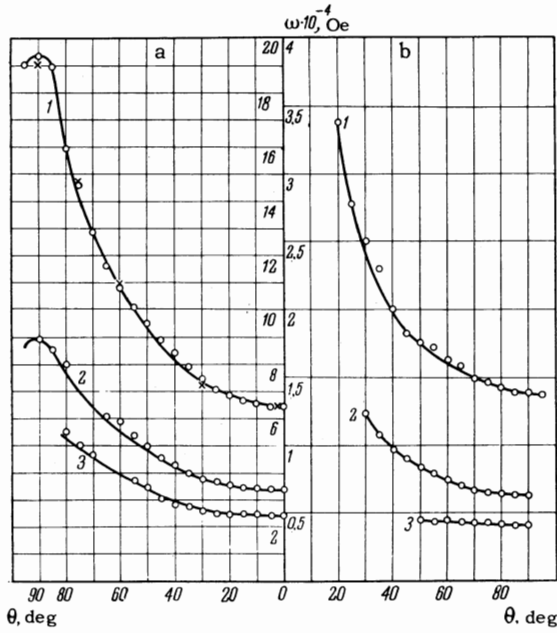


FIG. 4. Angular dependence of the frequency  $\omega$  of the electrical resistance oscillations, associated with the hole (a) and electron (b) parts of the Fermi surface of bismuth, recorded at various pressures;  $\circ$  – results of the present investigation,  $\times$  – results of Brandt et al.<sup>[7]</sup> (taken, for the sake of illustration, from a smoothed-out curve); continuous curves: a) calculation for the ellipsoidal model. 1)  $P = 1$  bar; 2)  $P = 11$  kbar; 3)  $P = 15$  kbar.

confirms the tendency of the hole part of the Fermi surface of bismuth to become spherical under pressure, as suggested by Brandt.<sup>[8]</sup>

Using the results obtained for the hole ellipsoid, we can calculate the change in its volume under pressure:

$$\frac{V(P)}{V_0} = \frac{S_{\max}(P)}{S_{0\max}} \left[ \frac{S_{\min}(P)}{S_{0\min}} \right]^{1/2} = 0.33 \pm 0.05$$

for  $P = 11$  kbar,

$$V(P)/V_0 = 0.2 \pm 0.05 \quad \text{for } P = 15 \text{ kbar.}$$

We were unable to investigate experimentally the anisotropy of the electron surface deformation because we did not measure the largest cross sections of the electron ellipsoid even at atmospheric pressure. The anisotropy of the deformation of the electron ellipsoid by pressure can be calculated assuming equality of the volumes of the hole and electron ellipsoids. The volume of the electron ellipsoid is  $V \sim (S_1 S_2 S_3)^{1/2}$ , where  $S_1, S_2, S_3$  are the areas of the principal cross sections of the ellipsoid ( $S_1$  is the smallest cross section). Assuming that the ratio of the areas  $S_2/S_3$  is independent of pressure, we find that

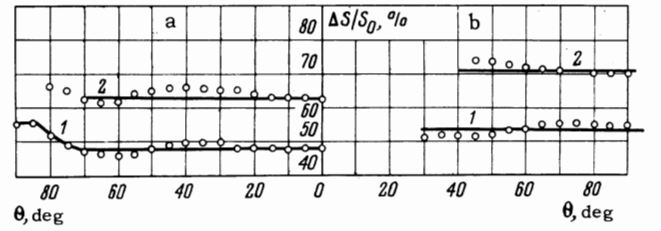


FIG. 5. Angular dependence of the relative change in the extremal cross sections of the hole (a) and electron (b) ellipsoids at various pressures: 1)  $P = 11$  kbar; 2)  $P = 15$  kbar.

$$\frac{S_3(P)}{S_3^0} = \frac{V(P)}{V_0} \left( \frac{S_1^0}{S_1(P)} \right)^{1/2}$$

Using the experimental data

$$V(11 \text{ kbar})/V_0 = 0.33 \pm 0.05,$$

$$S_1(11 \text{ kbar}) = (0.47 \pm 0.04) S_1^0,$$

we find that  $S_3(11 \text{ kbar}) = (0.5 \pm 0.1) S_3^0$  and

$$(S_3/S_1)_{11 \text{ kbar}} = (1 \pm 0.3) (S_3/S_1)_0.$$

Thus, the anisotropy of the electron ellipsoid is not greatly affected by a pressure of 11 kbar.

It should be mentioned that at high pressures (15–20 kbar) the period of oscillations associated with the small cross sections of the electron surface increases so much that only two or three oscillations can be observed in the whole range of magnetic fields and the accuracy of the determina-

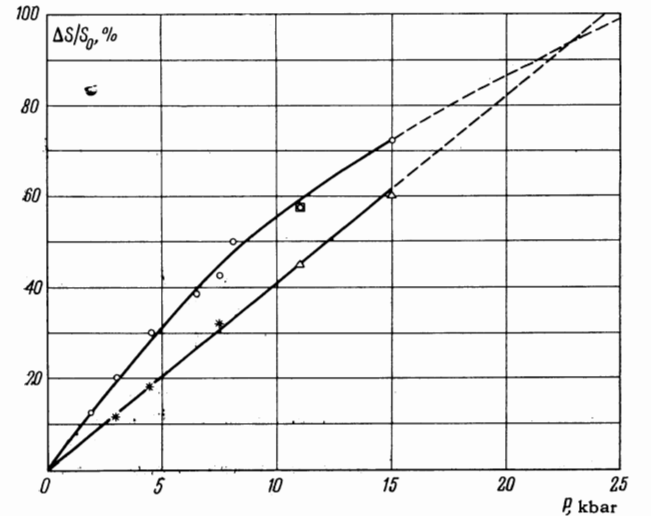


FIG. 6. Pressure dependence of the relative change in the area of the extremal cross sections of the electron and hole ellipsoids:  $\circ, \Delta$  – results of the present investigation for the small cross sections of the electron and hole ellipsoids;  $\square$  – ditto, for the large cross sections of the hole ellipsoid;  $\bullet, *$  – results reported in <sup>[2]</sup> for the small cross section of the electron and hole ellipsoids.

tion of the areas of these cross sections (for the purpose of estimating the anisotropy) is low. The considerable increase in the period of the electron oscillations at angles  $\theta$  close to  $90^\circ$  makes it difficult to investigate the deformation of the Fermi surface of bismuth at pressures close to a possible electron transition and, therefore, it would be desirable to observe the quantum oscillations associated with the hole ellipsoid at pressures of 15–20 kbar.

The results reported in<sup>[2]</sup>, obtained at pressures up to 8 kbar, and those reported in the present paper for pressures up to 15 kbar are used in Fig. 6 to plot the pressure dependence of the relative change in the small cross sections of the electron and hole ellipsoids. For the small hole cross sections,  $\Delta S/S_0$  depends linearly on the pressure and  $\Delta S/S_0 P \approx 4\%/kbar$ . The relative change in the small electron cross sections depends nonlinearly on the pressure. Extrapolating the obtained dependences of  $\Delta S/S_0$  on  $P$  in the direction of higher pressures, we find that the points at which the small cross sections of both ellipsoids may possibly vanish approximately coincide, and, consequently, the postulated electron transition lies at a point in the pressure range 24–26 kbar.

In conclusion, the authors regard it as their pleasant duty to thank Academician L. F. Veresh-

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