Investigation of bismuth in a quantizing field

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Quantum oscillations of the conductivity of bismuth located in fields up to 65 kOe and possessing a temperature 0.4°K and the velocities of magnetoplasma waves with frequencies 17.60 and 27.87 GHz are investigated at a frequency of 10 MHz, the field **H** being directed along the bisector and trigonal axes. The field dependences of the current carrier density and of the Fermi energy are measured. Values are obtained for the energy gap ($E_g^h = 0.7^{+0.7}_{-0.3}$ eV) at the point T, the hole cyclotron mass at the top of the valence band [$m_0 = (0.205 \pm 0.003 m_e$], and the electron Fermi energy ($E_F e = 20 \pm 3$ meV). The bottom of the electron energy band as determined by the position of the Landau level with $n = 0$ and $s = -1/2$ is found to decrease with increasing field intensity. The change is 0.05 ± 0.025 meV/kOe. It is shown that the ratio of spin cyclotron splitting for holes is less than 0.05 at $\mathbf{H} \perp C_3$ and is 2.13 for $\mathbf{H} \parallel C_3$. The decrease of the energy corresponding to the bottom of the electron band with increasing field and the large spin splitting of the Landau hole levels contradict the predictions of the deformation theory in which allowance is made for the interaction between four bands at the points L and T.[4,7] The dielectric-tensor components of the lattice measured in the microwave band (are $\epsilon_\parallel = 65 \pm 5$) along the C_3 axis and $\epsilon_\perp = 100 \pm 10$ perpendicular to the C_3 axis.

When a magnetic field $H \gtrsim 15-30$ kOe is applied to bismuth, there remains a single Landau level in each of the electronic conduction bands below the Fermi level E_F. The electron concentration at these levels, at $T = 0°K$, is [1]

$$N = 2eHp_z/(2\pi\hbar)^2 c \qquad (1)$$

(p_z is the maximum momentum of the electrons in the field direction), and therefore when the field is increased the equality of the number of electrons and holes is preserved by shifting E_F in such a way that p_z is decreased. The energy increments caused by fields of ~100 kOe are small in comparison with atomic increments, and do not change the band structure [1]. The influence of the field on the band structure can be estimated from the change of the latter following application of a uniaxial stress equal to the magnetic pressure. In fields ~100 kOe the susceptibility of bismuth is $\sim 3 \times 10^{-5}$ [2], and the pressure of the magnetic field on the crystal is equal to $\sim 10^5$ dyn/cm^2. When such a pressure is applied, the extremal sections of the Fermi surface of the electrons change in accordance with [3] by $\delta S/S \approx 10^{-4}$.

Thus, an investigation of bismuth in strong fields offers a unique possibility of experimentally studying the carriers at different values of the limiting energy E_F. This problem is of interest also because so far the origin and structure of the electron spectrum in bismuth are not yet clear [4,5,6]. At the same time, the results of [7,8], where the energy levels in a quantizing field were calculated, make it possible to connect the experimental data with the possible models of the spectrum.

In bismuth, in fields ~100 kOe and higher, quantum oscillations (QO) of the conductivity were investigated earlier [9,10] and the velocities of the magnetoplasma (Alfven) waves (MPW) were measured [11-13]. The comparison of the results of these papers shows that they are in poor agreement and in some cases contradict one another (for example [12] and [13]). Since the studies in [9-13] were made on the same experimental level, it was possible to ascertain the true behavior of bismuth in a quantizing field only by performing measurements at accuracies higher by one order of magnitude.

Contributing to the increase in the measurement accuracy was the improvement in the procedure for preparing the bismuth single crystals [14] and the production of samples having carrier relaxation times $\tau \approx 2-5$ nsec [15]. In the investigation of crystals of this type, it was deemed advisable to lower the temperature to ~0.1°K, and this made it possible to measure all the extremal sections of the Fermi surface of bismuth with accuracy ~0.1–0.2% [16]. Preliminary investigations of the QO and the MPW in a quantizing field have shown that the wave velocity can be measured with the same accuracy [17].

In the present study, we investigated the QO and MPW in fields up to 65 kOe. A simultaneous investigation of these effects made it possible to determine independently N(H) and E_F(H) [17]. The advantages in measuring QO and MPW in the same samples during the same experiments are undisputed. This eliminates many factors, sometimes difficult to control, that make the comparison of the measurement results difficult.

We describe below the organization of the experiment and the results of the investigations. A comparison is made with different experiments and with the theory. A number of parameters of the bismuth spectrum are obtained or determined more accurately, and it is shown that the existing theory calls for improvement.

EXPERIMENT

The experiments were performed in fields up to 65 kOe on bismuth single crystals cooled to 0.4°K by pumping off ^{3}He vapor. We measured the quantum oscillations of the quasistatic conductivity at ~10 MHz [16] and the velocity of the magnetoplasma waves [18] in the frequency range f = 18–30 GHz.

The bismuth single crystals were grown in a dismountable quartz mold by a method described earlier [14]. The samples were disks of 18 mm diameter and ~2 mm thickness. For one sample, the orientation of the normal **N** coincided within ~1° with the direction of the C_2 axis, and for the other the angle between **N** and C_3 was 3° 45′ ± 15′. (C_1, C_2, and C_3 are respectively the bisector,

binary, and trigonal axes). The same samples were used earlier to investigate the quantum oscillations at $T \approx 0.1°K$ [16], and it was established that the single temperature of these samples is $\lesssim 0.1°K$.

The sample thickness was determined with accuracy ~0.002 mm by weighing and by measuring the diameter. The correction for the thermal expansion when cooling to low temperatures was calculated from the data of White [19].

Measurement of the MPW velocity. The low-temperature part of the apparatus is shown schematically in Fig. 1. The sample served as a wall for a cylindrical resonator of 15 mm diameter and 6 mm height. At the center of the resonator was placed a strip of length ~6.5 mm. The natural frequency of the resonator in the TEM mode was 17.6 GHz. Volume-mode oscillations at frequencies ~25 and 28 GHz could also be excited in the resonator.

Changing the magnetic field changed the velocity of the MPW, leading to a periodic satisfaction of the Fabry-Perot resonance condition in the bismuth plate at

$$r\lambda/2=d, \tag{2}$$

where r is an integer, d is the sample thickness, and λ is the wavelength in the metal, $\lambda = 2\pi/k \ll \lambda_0 = c/f$. This is accompanied by a change in the amplitude of the microwave field at the bolometer. The derivative of the bolometer signal was registered by the known modulation method.

The width of the resonances was determined by the wave damping, which was equal to $1/2\omega\tau$, [18] and by the radiation losses, which are proportional to the refractive index λ/λ_0. With increasing field, λ increases, and in strong fields, for high-grade samples, the relative width of the Fabry-Perot resonances increases strongly as a result of the radiation losses.

The increase of λ/λ_0 exerts a particularly strong influence on the observed picture of the oscillations, when the sample serves as a wall of a measuring cavity with $Q \gtrsim 10^3$ (Fig. 2, lower curve). In this case, at $\lambda/\lambda_0 \gtrsim 10^{-2}-10^{-3}$, the sample and the cavity turn out to be strongly coupled. As a result, as shown by calculation, the bolometer registers strongly broadened MPW resonances at field values corresponding to the condition

$$(2r+1)\lambda/4=d. \tag{3}$$

In the experiment, the condition (2) corresponds to weak fields, while condition (3) corresponds to extremely strong fields, while in the intermediate range of fields (corresponding to Fig. 2) it is difficult to calculate the shift of the resonances. To eliminate the shift and the broadening of the resonances, it was necessary to decrease the coupling between the sample and the resonator. To this end, an aluminum film ~0.1 μ thick (on the order of the depth of the skin layer) was evaporated on the surface of the sample in vacuum. A film of aluminum ~0.1–0.5 μ thick was evaporated on the cylindrical surface of the sample to suppress the MPW resonances, which have a characteristic dimension of the order of the sample diameter [18]. After evaporating the aluminum film, the amplitude of the resonance decreased by several orders of magnitude, but they narrowed down significantly to a relative width $1/2\omega\tau \approx 1/200-1/400$ and, as shown by calculation, their position in this case was determined by formula (2) (Fig. 2, upper curve).

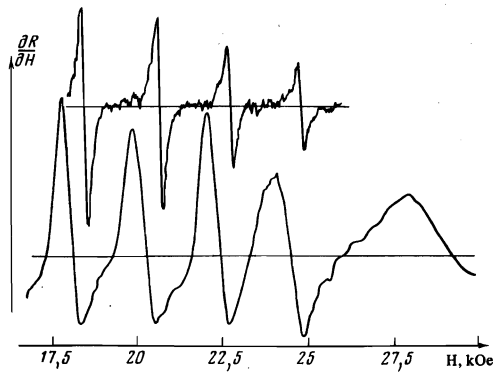

FIG. 2. Plots of the derivative of the bolometer resistance at 27.87 GHz; $\mathbf{H} \parallel C_1$, $\mathbf{k} \parallel C_2$. The upper curve was obtained with the sample coated with an aluminum film ~0.1 μ thick, and the lower without the film; T = 0.4°K.

The MPW-velocity measurement accuracy determined by the resonance width amounted to ~0.1% in the entire employed field range 5–65 kOe.

Registration of quantum oscillations of the conductivity. The coil of a 10-MHz tank circuit was placed near the sample on the side outside the resonator. The tank circuit was connected in a self-oscillator circuit with enhanced feedback. The feedback maintained the oscillation amplitude at the level ~10–100 mV, the power dissipated in the tank circuit being $\sim 10^{-6}-10^{-8}$ W. The magnetic field was varied and a signal proportional to generated-frequency deviation due to the magnetic-field modulation was registered. A typical plot of the quantum oscillations is shown in Fig. 3.

In strong fields, the depth of penetration becomes larger than the sample thickness, its change has little effect on the inductance of the tank circuit, and this leads to a decrease of the signal amplitude. Simultaneously, with decreasing signal, a change takes place also in the waveform of the QO line (Fig. 3). This can be due in part to the fact that the contribution to the frequency deviation from the susceptibility oscillations, which is proportional to $\partial M/\partial H$, becomes comparable with the contribution from $\partial R/\partial H \propto \partial^2 M/\partial H^2$.

The aluminum film coated on the sample altered the observed picture strongly. Thus, for a sample without the film, the signals from the holes and electrons were comparable in amplitude but of opposite sign at $\mathbf{H} \parallel C_1$ and $\mathbf{N} \parallel C_3$. After coating the sample on all sides with an aluminum film, whose thickness ~0.1 μ was much less than the depth of the skin layer at 10 MHz, the sign of the signal for the electrons became the same as for the holes, and the signal amplitude remained of the same order of magnitude.

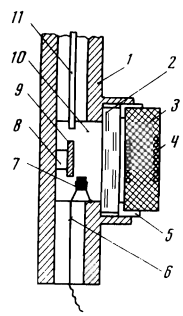

FIG. 1. Low-temperature part of the apparatus: 1–housing of resonator; 2–sample, 3–epoxy compound, 4–coil of 10-MHz tank circuit, 5–ring, 6–bolometer lead, 7–carbon bolometer, 8–foamed polystyrene, 9–resonant strip line, 10–resonator cavity, 11–central conductor of coaxial lead.

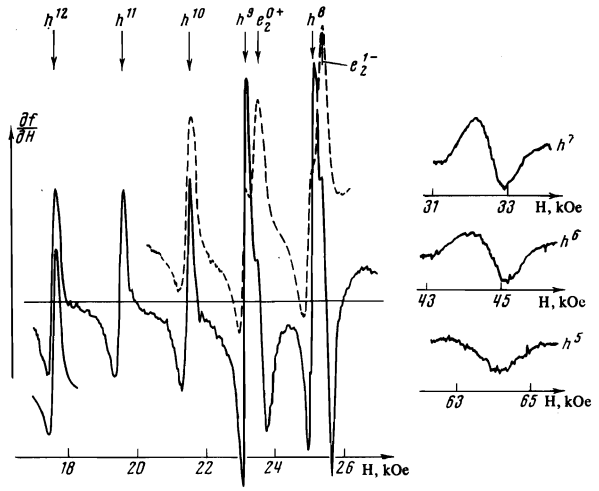

FIG. 3. Plot of the quantum oscillations of the conductivity at 10 MHz and T = 0.4°K; $\mathbf{H} \parallel C_1$, $\mathbf{N} \parallel C_3$. The solid curve was obtained with a sample without a film, and the dashed curve with a sample coated with an aluminum film ~0.1 μ thick. The calculated line shape is shown under the curve for ~18 kOe. On the top are indicated the orders of the oscillations of the holes (h) and of the electrons of the inclined ellipsoids (e_2) at different spin directions.

In a field $H \lesssim 30$ kOe, the waveform of the line (Fig. 3) was close to the calculation results for $\partial^2 M/\partial H^2$ [20] with the same parameters which were used by us earlier [16]. In these cases it was possible to determine the value of the field H_n corresponding to tangency of the n-th Landau level of the Fermi surface, with accuracy ~ 0.1%. At $H > 30$ kOe, it is impossible to calculate the line shape, since we did not know a number of parameters that determine the electrodynamics of the problem, for example the ratio of the skin-layer depth and the sample dimensions. In this case H_n was taken to be the field corresponding to the center of gravity of the line. The resultant error could then increase to ~0.5%.

Magnetic field. The field ~65 kOe produced by the solenoid was homogeneous inside the sample to within not more than 10^{-4}. The solenoid constant was determined in the field range 10–50 kOe with accuracy ~0.02% by NMR calibration of the protons with a running-water nuclear magnetometer. To monitor the field, DPPH was placed in the resonator during the experiments, and the EPR signal was observed at the frequencies 17.6 and 28 GHz. From the EPR signals and from the QO and MPW spectra it was established that the field changed in reproducible fashion with accuracy better than ~0.1%, and its values were in agreement with the expression

$$H[\text{Oe}] = 464J[\text{A}] - 20. \qquad (4)$$

The solenoid was mounted in a universal joint and could be inclined during the experiment in two perpendicular planes to an angle ±4° with accuracy ~0.5′. The field was set parallel to the chosen bismuth-crystal axis with accuracy ~50′, by using the QO signal.

A modulating field with amplitude up to ~50 Oe and frequency 6.25 Hz, synchronized with the mains voltage, was applied parallel to the constant field. Although in strong fields the line width exceeded this value by one order of magnitude, it was impossible to increase the amplitude of the modulation field, for otherwise overheating of the sample would lead to a broadening of the QO lines.

EXPERIMENTAL RESULTS

1. Quantum Oscillations

Field $\mathbf{H} \parallel C_1$. Figure 3 shows a plot of the QO at $\mathbf{H} \parallel C_1$. The measured values of H_n were used to determine the carrier concentration. In the calculations it was assumed that the hole spectrum is described by a two-band model corresponding to the energy levels [21]

$$E^h(1+E^h/E_g^h) = (n+1/2)\hbar\Omega + p_z^2/2m_1, \qquad (5)$$

where E_g^h is the gap between the bands; E^h is the energy reckoned downward from the top of the valence band; $\Omega = eH/m_{01}c$; p_z is the momentum along the field $\mathbf{H}$; m_{01} and m_1 are the masses at the top of the band.

Recognizing that the oscillations are observed at field values corresponding to $p_z^{(n)} = 0$ in (5), we can obtain in accordance with (1), after summing p_z for all the Landau levels with $k < n$,

$$\frac{N(H)}{N(0)} = \frac{3}{2}(H_n \Delta H^{-1})^{3/2} \sum_{k=0}^{n} (n-k)^{1/2}, \qquad (6)$$

where $N(0) = 3.015 \times 10^{17}$ cm^{-3}, $\Delta H^{-1} = 4.717 \times 10^{-6}$ Oe^{-1} is the period of the oscillations in the reciprocal field [16], and H_n is the field corresponding to the quantum number n. The absolute values of n were determined earlier in the quasiclassical field region [16]. Plots of N(H)/N(0) are shown in Figs. 4 and 5. Formulas (5) and (6) are valid if there is no spin splitting of the Landau levels. The spin splitting of the hole level at $\mathbf{H} \parallel C_1$ can be estimated by measuring the QO in a field directed at an angle ϑ to the basal plane. With increasing ϑ, the amplitude of the oscillations decreases. At $\vartheta \gtrsim 2°$, each peak splits into two and the spin splitting in the reciprocal field ΔH_{sp}^{-1}, increases in proportion to ϑ. On the basis of these experiments, and from an analysis of the QO line shape, it was established that at $\mathbf{H} \parallel C$ we have a ratio $\Delta H_{sp}^{-1}/\Delta H^{-1} \lesssim 5\%$. The possible error in the determination of N(H) is in this case less than $7n^{-3/2}\%$, which amounts to $\lesssim 0.7\%$ at n = 5 (H = 64.1 kOe).

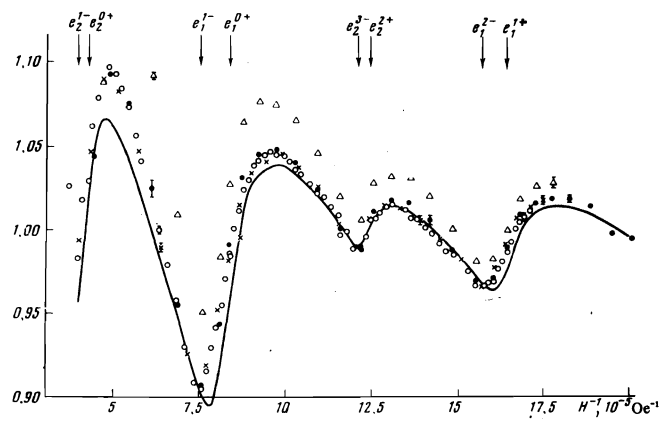

FIG. 4. Field dependence ($\mathbf{H} \parallel C_1$) of the following relative values: X–hole density N(H)/(H(0), O–mass density $NF_{22}^1(H)/NF_{22}^1(0)$ determined from the MPW velocity and •–$NF_{33}^1(H)/NF_{33}^1(0)$; Δ–values of $NF_{33}^1(H)/NF_{33}^1(0)$ without allowance for $\epsilon_3 = 65$. Microwave oscillation frequency 27.87 GHz. The arrows indicate the position of the electron oscillations for an ellipsoid whose major axis lies in the bisector plane parallel to the field (e_1), and for inclined ellipsoids (e_2). The upper index indicates the number of the Landau level in the spin direction. The solid curve gives the calculated N(H)/N(0) dependence in accordance with the Lifshitz-Kosevich theory.

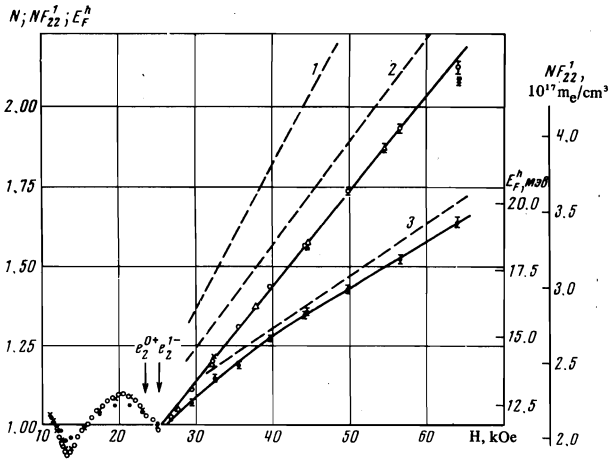

FIG. 5. Dependence on the field of the following relative quantities: X–N(H)/N(0); ○, △–$NF^1_{22}(H)/NF^1_{22}(0)$ respectively at f = 27.87 and 17.6 GHz; ●–$E^h_F(H)/E^h_F(0)$. The dashed curves 1 and 2 show the values of $NF^1_{22}(H)/NF^1_{22}(0)$, obtained respectively in [12] and [11]; curve 3–from [9]. The right-hand scale pertains only to the data of the present paper.

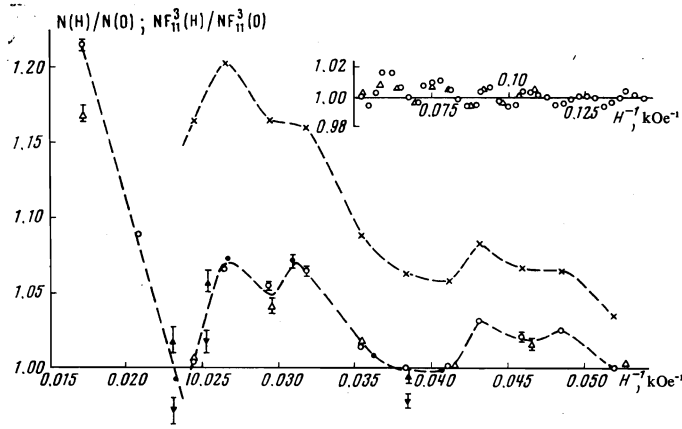

FIG. 6. Field dependence at $\mathbf{H} \parallel C_3$ and $\mathbf{N} \parallel \mathbf{k} \parallel C_2$ of the following quantities: △–electron concentration, ○–value of NF^3_{11} at f = 27.87 GHz, ●–at f = 17.6 GHz, and X without allowance for $\epsilon_1 = 100$, and also of the hole density at $\Delta H^{-1}_{sp}/\Delta H^{-1} = 2.13$ and 1.87 (the points ▲ and ▼, respectively).

Allowance for the finite temperature T = 0.4°K can yield a correction on the order of 2/n%. Neither error exceeds the error connected with the measurement of H_n (~0.5–0.1%) and H^{-1} (0.1–0.2%). Thus, the accuracy with which N(H) is measured is 1–0.5%.

Field $\mathbf{H} \parallel C_3$. We present below the values of H_n for QO of electrons and holes:

	n:	0	1	2	2	3	3	4	4	5	5
	s:	$+^1/_2$	$+^1/_2$	$-^1/_2$	$+^1/_2$	$-^1/_2$	$+^1/_2$	$-^1/_2$	$+^1/_2$	$-^1/_2$	$+^1/_2$
H, kOe	electrons:		58.5	40.9	33.8	28.17	24.06	21.46	18.96	17.24	15.66
	holes:	39.5	24.1	43.3	17.66	26.0		18.3			

At this field direction, to determine the change of the carriers it is more convenient to use the QO for the electrons, since they are encountered approximately three times more frequently. However, taking into account the large spin splitting, it is necessary to use a modified two-band model [9], which differs from (5) in that n + 1/2 is replaced by

$$n+{}^1/_2 \pm \Delta H_{cn}{}^{-1}/2\Delta H^{-1}.$$

In this case $\Delta H^{-1}_{sp}/\Delta H^{-1} = 0.54 \pm 0.01$ [18]. In this approximation we can obtain a formula analogous to (6) for the calculation of N(H)/N(0); the dependence of this ratio on the field is shown in Fig. 6.

2. Magnetoplasma Waves

At $\mathbf{H} \parallel C_1$ and $\mathbf{H} \parallel C_3$, in strong fields, the off-diagonal components of the conductivity tensor of pure bismuth are equal to zero, and at $\mathbf{k} \perp \mathbf{H}$ the connection between the wave spectrum and the carrier spectrum is given by

$$k^2 - \varepsilon_\alpha \frac{\omega^2}{c^2} - \frac{4\pi i}{c}\omega\sigma^j_{\alpha\alpha} = 0, \qquad (7)$$

where α = 1, 2, 3 corresponds to different polarizations of the wave relative to the crystal axes (the electric field of the wave is parallel to the axes C_1, C_2, and C_3, respectively), j is the direction of the field, $\epsilon_1 = \epsilon_2 \neq \epsilon_3$ are the components of the dielectric tensor of the lattice, $\sigma^j_{\alpha\beta}$ are the components of the conductivity tensor, and $\omega = 2\pi f$. In strong fields, when $\Omega = eH/m^*c \gg \omega$, $kv \ll \omega$ (v is the carrier velocity), the conductivity is

$$\sigma^j_{\alpha\alpha} = \frac{c^2\omega}{iH^2} NF^j_{\alpha\alpha}(m). \qquad (8)$$

for an isotropic spectrum, $F^j_{\alpha\alpha}(m) = m_e + m_h$. It is easy to verify that for the conductivity given by formula (8), the wave velocity $V = \omega/k$ does not depend on ω.

Formula (8) was obtained in the classical limit; it was shown in [22] that in the case of a quadratic isotropic spectrum of the carriers the conductivity tensor in the quantum limit, in the absence of dispersion, retains its form:

$$\sigma_{\alpha\alpha} = \frac{c^2\omega}{iH^2}\sum_{e,h} Nm.$$

at $\mathbf{H} \parallel C_1$, the main contribution to $\sigma^1_{\alpha\alpha}$ is made by holes having a spectrum close to quadratic. Using (7), we can determine $NF^1_{\alpha\alpha}(H)$ from the measured values of k.

The value of k was determined in accordance with formula (2). The absolute values of the orders of the resonances r were chosen such that the functions V(H), measured at different frequencies f = 17.6, 25.17, and 27.87 GHz, coincided in best fashion. Owing to the small width of the resonance lines (~0.2–0.5%) and the relatively small $r \approx 10$–100, the value of r was determined uniquely. At the chosen values of r, the functions V(H) coincided, within the measurement accuracy, in the field region H ≈ 10–20 kOe.

In strong fields, we observed a velocity difference that reached ~2% at $\mathbf{H} \parallel C_3$ in the maximum field 65 kOe. This difference is connected, in our opinion, with the presence of impurities in the bismuth. The impurities produce a difference between the electron and hole densities. Consequently, off-diagonal components of the conductivity tensor $\sigma_{\alpha\beta} \propto (\Delta N/N)\Omega\sigma_{\alpha\alpha}/\omega$ appear in strong fields, and $\sigma_{\alpha\alpha}$ in (7) must be replaced by $\sigma_{\alpha\alpha} - \sigma^2_{\alpha\beta}/\sigma_{\beta\beta}$. From the difference in the wave velocities at f = 17.6 and 28 GHz we obtained an estimate $\Delta N/N(0) \approx (0.4 \pm 0.2)\%$ for the sample with the normal along C_3. For the sample with the normal along C_3 the measured value at $\mathbf{H} \parallel C_3$ was $(\Delta N/N(0) = (0.1 \pm 0.02)\%$.

At $H \lesssim 10$ kOe it is necessary to take into account the temporal and spatial dispersions. The corrections to the wave vector can be calculated from the formulas given in [23]. The wave velocity in the presence of dispersion determines not only $\sigma^j_{\alpha\alpha}$, but also the other conductivity tensor components, $\sigma^j_{\alpha\beta}$ and $\sigma^j_{\beta\beta}$. Calculations have

shown that the frequency-dependent corrections to V frequency are proportional to H^{-2} and do not exceed ~1% at H ≈ 6 kOe. After introducing these corrections, the values of V(H) obtained at different ω turn out to be equal within the measurement accuracy also at H ≲ 10 kOe. In this field region, the contribution of ϵ_α to the wave velocity is small, and the values of $NF^j_{\alpha\alpha}(H)$ obtained from (7) at $\epsilon_\alpha = 0$ practically coincide with the values of N(H) (Figs. 4 and 6); this serves as an additional confirmation of the correct choice of the orders of the resonance r.

With increasing field, the contribution of ϵ_α increases and the values of $NF^j_{\alpha\alpha}(H)$, calculated from the measured values of k without allowance for ϵ_α, turn out to be much larger than N(H). This difference has made it possible to determine the components of the dielectric tensor of the bismuth lattice. The values of ϵ_2 and ϵ_3 were chosen at $\mathbf{H} \parallel C_3$ and $\mathbf{H} \parallel C_1$, $\mathbf{k} \parallel C_2$ in such a way that the corresponding values of $NF^j_{\alpha\alpha}(H)/NF^j_{\alpha\alpha}(0)$ coincided with the values of N(H)/N(0) in that range of fields for which this ratio differed little from unity (see Figs. 4 and 6). We thus obtained $\epsilon_2 = 100 \pm 10$ and $\epsilon_3 = 70 \pm 10$.

In strong fields at $\mathbf{k} \parallel C_2$ and $\mathbf{H} \parallel C_1$, the contribution made to k by ϵ_3 and NF^1_{33} are comparable in magnitude, so that the value of ϵ_3 can be made more precise by assuming that expression (8) remains valid also in the quantum region for a nonparabolic carrier spectrum. The relative contribution of the electrons to NF^1_{33} in weak fields is equal to ${}^eNF^1_{33}/NF^1_{33} = 22.5\%$, and the contributions of the electron ellipsoid, the major axis of which is parallel to H, and of the two others have a ratio 1 : 4 : 4. In a field H ≳ 26 kOe, the number of carriers in each inclined ellipsoid becomes half as large than in the parallel ellipsoid, and this ratio changes to 1 : 2 : 2. Recognizing that the total electron density remains equal to the hole density, we find that in the expression that determines ${}^eNF^1_{33}$ there appears an additional factor (3/2)(5/9) = 5/6. The relative contribution of the electrons becomes equal to 19.5% and with further increase of the field it decreases in proportion to the decrease of the effective mass, owing to the decrease of the Fermi energy.

A plot of ${}^eNF^1_{33}/NF^1_{33}$ is shown in Fig. 7. To construct this plot, NF^1_{33} was determined from the measured value of c^2k^2/ω^2, from which we subtracted $\epsilon_3 = 65$. The quantity ${}^eNF^1_{33}$ was assumed to be the difference $NF^1_{33} - {}^hNF^1_{33}$. The contribution of the holes to NF^1_{33} at this orientation is equal to ${}^hNF^1_{33} = (S_3/S_1)^2\,{}^hNF^1_{22}$, where $S_3 = 6.76 \times 10^{-42}$ g²cm²/sec² and $S_1 = 22.49 \times 10^{-42}$ g²cm²/sec² are respectively the minimal and maximal sections of the Fermi surface of the holes [16], and ${}^hNF^1_{22} = 0.985\ NF^1_{22}$.

According to Fig. 7, the change of ${}^eNF^1_{33}/NF^1_{33}$ with increasing field corresponds to the predicted value at $\epsilon_3 = 65 \pm 5$. This value of ϵ_3 agrees within the limits of errors with that given above, but it is determined with double the accuracy.

DISCUSSION OF RESULTS

1. Boyle and Brailsford [24] measured $\epsilon_1 = 100$ in the infrared band at $\omega > \omega_{pl}/\sqrt{\epsilon}$ (ω_{pl} is the plasma frequency); this agrees with our value 100 ± 10. The value of ϵ_3 can be determined from the ratio of the frequencies corresponding to the boundaries of the plasma absorption for waves polarized along and across the trigonal axis. Using the results of [24] and calculating the components of the conductivity tensor which determines ω_{pl}, we obtain from the bismuth-spectrum parameters given in [16,25] the value $\epsilon_3/\epsilon_1 = 0.71 \pm 0.03$, i.e., $\epsilon_3 = 70 \pm 10$. Our measured value is $\epsilon_3 = 65 \pm 5$. Thus, there is no dispersion of the dielectric constant of the bismuth lattice in a wide range of frequencies (from 10^{10} to 10^{13} Hz). This conclusion agrees with Abrikosov's theory [26].

2. Table I compares the results of the measurements of $NF^j_{\alpha\alpha}$ with calculation based on the ellipsoidal model of the Fermi surface of electrons and holes, with parameters taken from [16,25]. For holes we used the cross section values given above, and the effective masses $(0.212 \pm 0.0005)m_e$ and $(0.0635 \pm 0.0005)r_e$, determined from the cyclotron resonance during the course of the present investigation. The differences between the quantities in the table do not exceed ~1% and correspond to the accuracy of the measurement and of the calculation. This proves that in bismuth there are no other carriers except those already known, one group of holes and three equivalent groups of electrons.

3. Using highly perfected and pure single crystals, we have established that there are uncompensated carriers with concentration 3×10^{14} cm^{-3}, which corresponds to an impurity concentration ~10^{-6}%. With such an unbalance, at f = 17.6 GHz in a field ~60 kOe, the value of NF^3_{11} turns out to be smaller by ~3% than the value for pure bismuth.

Since the influence of the impurity conductivity on the wave velocity increases in proportion to $(\Delta N\Omega/N\omega)^2$, it is clear that when experiments are performed in strong fields this circumstance must be seriously taken into account. In [11-13] they did not consider the role of the impurities. At the same time, one of the causes of the decrease of NF^3_{11} with increasing field, which has been observed in [13] and contradicts our results and the results of others [9,12], may be due precisely to this effect.

4. Figure 2 shows, besides the experimental points for N(H), also theoretical curves obtained by the Lifshitz-Kosevich formulas [20]. We used in the calculation the value $|\partial^2 S_1/\partial p_1^2|^{-1/2} = 6.41$ calculated for electrons in ac-

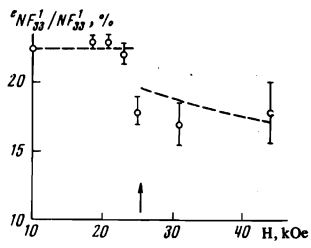

FIG. 7. Field dependence of the relative contribution of the electrons to $NF^1_{33}(H)$. The arrow indicates the value of the field above which only one Landau level remains in each of all the electron bands. Dashed line—calculation (without allowance for the quantum oscillations in fields H ≲ 25 kOe). The vertical lines next to the points indicates the error connected with the error in the determination of $\epsilon_3 = 65 \pm 5$.

TABLE I

	$NF^j_{\alpha\alpha}$, 10^{17} m_e/cm^3			
	$NF^1_{22}(0)$	$NF^1_{33}(0)$	$NF^3_{11}(0)$	$NF^2_{11}(0)$
Experiment	2.09±0.01	0.248±0.001	1.009±0.005	2.19±0.01
Calculation	2.115±0.02 *	0.248±0.002	1.00±0.01	2.17±0.02

*Calculated with correction for the deviation of the C_3 axis from the normal to the sample surface.

cordance with the model constructed in [16]. What is observed is not only good qualitative but also quantitative agreement between experiment and calculation. Small differences are connected mainly with the fact that no account is taken of the shift of the oscillation fields for electrons with change of the Fermi level.

Table II lists the values of the oscillation field with n = 0 and 1 at different field directions. The data at $\mathbf{H} \parallel C_2$ were obtained by us earlier [17]. If it is recognized that $\Delta H^{-1} = 2\pi e\hbar/cS$, and

$$S = S(0) + 2\pi m^* \delta E = S(0) - 2\pi m^* \frac{2}{3}\frac{\delta N}{N(0)} E_F^h(0),$$

where $\delta N/N(0)$ is determined directly from Fig. 4 (or from the figure in [17]), and if the suitable correction is introduced, then we find as a result that the phase of the oscillations retains the quasiclassical value $(n + \frac{1}{2} \pm \Delta H_{sp}^{-1}/2\Delta H^{-1}) = 1 \pm 0.045$ even at n = 0 or 1.

We note also that the contribution of the electrons to the conductivity tensor in the quantum region is determined by the classical expression with accuracy not worse than ~10% (Fig. 7).

5. At H > 25 kOe and $\mathbf{H} \parallel C_1$, when the electron is left with only one Landau level below the Fermi level, a monotonic nearly-linear increase of the carrier density sets in (Fig. 5). The function $NF_{22}^1(H)$ increases somewhat faster than N(H), and at H = 64 kOe we have

$$\frac{NF_{22}^1(H)}{NF_{22}^1(H)} \Big/ \frac{N(H)}{N(0)} = \frac{F_{22}^1(H)}{F_{22}^1(0)} = 1.02 \pm 0.01.$$

The deviation of the presented ratio from unity makes it possible to estimate the deviation of the spectrum from quadratic.

The simplest among the possible spectra that take into account terms proportional to E^2 in the expansion of $p^2(E)$ is the spectrum of the two-band model

$$\hat{p\alpha}p = E(1 + E/E_g), \tag{9}$$

which corresponds to the energy levels (5) ($\hat{\alpha}$ is the inverse effective-mass tensor). The peculiarity of the spectrum (9) is that all the equal-energy surfaces are ellipsoids with equal axis ratio.

Using the definition of the effective mass, we can write down an expansion for the extremal section S in powers of E in the form

$$S = 2\pi E\left(m_0 + \frac{E}{2}\frac{\partial m_0}{\partial E}\right). \tag{10}$$

for holes, substituting the numerical values of S_1, S_3, m_1^*, and m_3^* from (10), which were given above, we obtain

$$\frac{S_1}{S_3}\Big|_{E=E_F^h(0)} = (1 \pm 0.01)\frac{m_1^*}{m_3^*} = (1 \pm 0.01)\frac{S_1}{S_3}\Big|_{E=2E_F^h(0)}.$$

The constancy of the ratio S_1/S_3 and the fact that the Fermi surface of the holes differs from an ellipsoid by not more than ~0.1—0.2% [16] justify the use of the spectrum (9).

In the model (9), the variation of m* as a function of the energy is described by the relation

$$m^* = m_0(1 + 2E/E_g^h). \tag{11}$$

Since $F_{22}^1(H)/F_{22}^1(0) = m^*(H)/m^*(0) = 1.02 \pm 0.01$, it follows that, using (11), we get $E_g^h = 0.7^{+0.7}_{-0.3}$ eV.

Knowledge of E_g^h makes it possible to find the mass on top of the band $m_{01} = (0.205 \pm 0.003)m_e$ as well as the Fermi energy $E_F^h(0) = 11.76 \pm 0.25$ meV and to construct the plot of $E_F^h(H)$—see Fig. 5. Figure 5 shows also a plot of $E_F^h(H)$, obtained by Smith et al. [9]. The proximity of their result to our value is accidental and is apparently due to averaging out of several errors, inasmuch as a comparison with our data has revealed that in fields ~20—25 kOe, where the peak of the large amplitude due to the electrons did not permit the authors of [9] to observe the hole oscillations, there are not one but two hole peaks. As a result the values of the quantum number n assumed in [9] at H < 20 kOe were smaller by unity than the true value.

TABLE II

n	s	H, kOe	$H\Delta H^{-1}$ *	$H\Delta H^{-1}$ **
		$\mathbf{H} \parallel C_1$		
1	−1/2	25.31	1.033	1.030
0	+1/2	23.46	0.955	0.967
1	−1/2	13.30	1.084	1.041
0	+1/2	11.96	0.975	0.966
		$\mathbf{H} \parallel C_2$		
1	−1/2	15.43	1.086	1.055
0	+1/2	13.95	0.982	0.973

*The error of the presented value is ~0.5%.

**With allowance for the change of E_F.

6. At $\mathbf{H} \parallel C_3$, the ratio $NF_{11}^3(H)/N(H)$ should change in proportion to $m^*(H)/m^*(0) = 1 + 2\delta E/E_{ph}$, where δE is the change of E_F^e, and $E_{ph} \approx 70$ meV is the photon energy corresponding to the light absorption-region edge connected with the interband transitions [27]. Using the results as shown in Fig. 6 for the estimate of $\delta E \propto (2/3)\delta N$, we find that $NF_{11}^3(H)/N(H) = 1.055$ at H = 64 kOe, and the experimentally measured ratio is 1.040. Thus, our procedure of calculating N(H) for electrons leads to an error not larger than ~2%.

7. Figure 6 shows the values of the hole concentration calculated from the quantum-oscillation field for the relative spin splittings $\Delta H_{sp}^{-1}/\Delta H^{-1} = 1.87$ and 2.13. Although in both cases the concentration ^hN turned out to be close to ^eN, the value 2.13 leads to better agreement at $H^{-1} = 0.0385$ kOe^{-1}.

Since two hole peaks split in spin are observed in a field of ~40 kOe, and the average position of the hole oscillations corresponds to the law $H_n^{-1} = \Delta H^{-1}(n + 1/2)$, the only alternative for the spin splitting remains $\Delta H_{sp}^{-1}/\Delta H^{-1} = 0.13$. However, in this case the calculated values of ^hN turned out to be smaller by 20—25% than ^eN. Thus, in accord with the results of the measurements of the anisotropy of the spin splitting [9], the value 2.13 must be assumed for the latter.

Abrikosov [4] has shown that a spin splitting larger than unity is possible if at the point T of the Brillouin zone, where the holes are located, there are closely-placed six rather than four energy levels. Thus, a large spin splitting points to the need of taking into account, in the determination of the parameters and of the form of the spectrum, of a larger number of levels than in [28].

8. It was shown in [7,8] that the level energy of the electron corresponding to the quantum numbers n = 0 and s = −1/2 changes linearly in a magnetic field. Beneslavskiĭ and Fal'kovskiĭ [7] obtained the dispersion law of this level. For the electrons of the ellipsoid whose major axis lies in the bisector plane parallel to the field, this dispersion law takes the form

$$p_1^2/2m_1 = E - aH, \qquad (12)$$

where the energy E is reckoned from the bottom of the band at H = 0. With four neighboring zones taken into account, it was shown that a > 0.[7]

An energy increment linear in the field appears in the theory when account is taken of the influence exerted on the conduction-electron spectrum by the bands at point L that are located ~1 eV away from the conduction band. Allowance for the more remote bands leads to a spin splitting of Landau levels with twofold degeneracy in the two-band model[7,8]. It becomes possible therefore to estimate the absolute value of a from the spin splitting of the electron levels[16]. Recognizing that the effective mass at the bottom of the band is equal to $m^*/(1 + 2E_F^e/E_g^e) \approx m^*/3$, using formula (26) from [7], we obtain a $\lesssim 0.1$ meV/kOe. A similar estimate can be obtained also from Baraff's theory.[8]

For inclined ellipsoids, the relative spin splitting remains unchanged, and the Landau splitting and p_z are practically half as large. It is therefore necessary to replace for them a in (12) by a/2, and m_1 by $m_1/4$ (more accurately, by $m_1/3.974$).

The function $p_1^2(E)$ can be easily obtained from experiment, by assuming that a = 0 and using formula (1), where the value used for $^eN(H)$ should be the one obtained for the holes. The change of the Fermi level is defined as $E_F^h(0) - E_F^h(H)$. This function, plotted in accordance with our data and the data of Brandt et al.[10], is shown in Fig. 8. The figure shows also the point obtained from [12] by introducing a correction to the wave vector, which leads to the agreement between NF_{22}^1 (64 kOe) with our data. The correction thus yields NF_{22}^1 (200 kOe) = 0.92 $\times 10^{18}$ $m_e cm^{-3}$. The initial section of the plot on Fig. 8, at $|\Delta E| < 10$ meV, is linear. This line, however, intercepts the ordinate axis at a point that is ~1% higher than the value of $p_1^2(0)$ of [16], and in strong fields the points do not fall on a straight line. These differences enable us to determine $E_F^e(0) = 20 \pm 3$ meV and $a = -(0.05 \pm 0.025)$ meV/kOe.

The value of $E_F^e(0) = 20$ meV is somewhat smaller than that obtained from magnetooptical investigations by reducing the experimental data in accord with the two-band model: $E_F^e = (E_{ph} - E_g^e)/2 \approx 26$–$28$ meV[27,29,30], or else from an analysis of the behavior of alloys, 32 ± 2 meV[31]. It seems that the difference is due to deviations from the two-band model, which leads, according to Beneslavskiĭ and Fal'kovskiĭ[2], to a renormalization of the effective mass on the Fermi level by an amount ~10%.

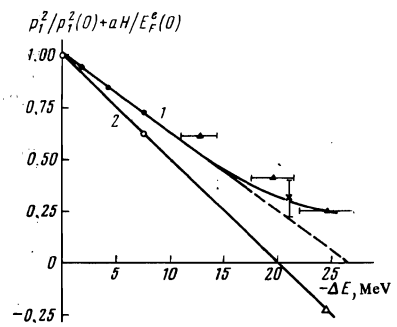

FIG. 8. Plot of $p_1^2/p_1^2(0) + aH/E_F^e(0)$ against $\Delta E = E_F^e(H) - E_F^e(0)$ at H ∥ C_1: ●, ○—obtained from the quantum oscillations in the present study, ▲, △—from [10], X—from [12], corrected in accordance with our results. Curve 1 was plotted under the assumption that a = 0, and curve 2 at a = −0.05 meV/kOe, $E_F^e(0) = 20$ meV.

The sign of a agrees with the anisotropy of the spin splitting, thus offering evidence that $\Delta H_{sp}^{-1}/\Delta H^{-1} > 1$[16] and is opposite to the sign predicted by the theory in [7]. This, as well as the already mentioned large spin splitting of the holes at **H** ∥ C_3, points to the need for taking into account a large number of bands in the theory.

9. From the carrier spectrum established in this paper, we can calculate all the thermodynamic quantities that are determined by the conduction electrons. Let us demonstrate this using as an example the calculation of the specific magnetic moment $M = -\partial\Omega/\partial H$. The thermodynamic potential Ω is determined by the known relation[1]

$$\Omega = -k_B T \sum \int_{-\infty}^{+\infty} \ln\left[1 - \exp\left(\frac{E_F - E(p_z)}{k_B T}\right)\right] n(p_z)\, dp_z. \qquad (13)$$

The summation in (13) is carried out over all the Landau levels and spin projections of the partially filled bands. Using the spectrum of the electrons and holes determined by formulas (12) and (5), and neglecting the small corrections $\sim E_F^h/E_g^h$, we obtain at **H** ∥ C_1 and T = 0

$$\Omega = -\frac{2}{3} N(H)\,[E_F^e(0) + E_F^h(0)] + \\ + \frac{4}{3}\frac{eH}{(2\pi\hbar^2)c}\left[p_1 aH + p_1' aH + 2\sum_{k=0}^{n} p_z^k \left(k + \frac{1}{2}\right)\hbar\frac{eH}{m_{01}c}\right], \qquad (14)$$

where p_1, p_1', and p_z^k are the limiting momenta respectively for the electrons of the straight and inclined ellipsoids and of the k-th Landau level for the holes.

Using the parameters of the bismuth spectrum determined in the present paper, and the plots of N(H) and $E_F^h(H)$ shown in Fig. 5, we obtain, with accuracy ~10%, for H in the range 30–60 kOe, the value $M = M_0 - \chi H = (0.19-0.3) \times 10^{-5} H$ [Gauss]. The value of M_0 is determined mainly by the first term of (14), and the contribution of the electrons to χ amounts to ~30%.

Direct measurements of the magnetic moment[2] yield $M_0 = 0.19$–0.30 G, and the contribution of the carriers to the diamagnetic susceptibility is $\sim(1-0.5) \times 10^{-5}$, which is close to the value obtained by us.

10. The increase of the carrier density with increasing field explains well the linear growth of the resistance obtained by Kapitza[32]. Indeed, at **H** ∥ C_1, the diagonal terms of the conductivity tensor in strong fields are $\sigma_{\alpha\alpha} \propto \sigma_{\beta\beta} \propto N(H)/\Omega^2\tau$. On the other hand, the hole components $\sigma_{\alpha\beta}$ are of the order of $\sigma_{\alpha\beta} \propto \Delta N/\Omega$. The experimentally measured resistivity ρ is expressed in terms of the conductivity tensor

$$\rho_{\alpha\alpha} = \sigma_{\alpha\alpha}^{-1}\left(1 + \frac{\sigma_{\alpha\beta}^2}{\sigma_{\alpha\alpha}\sigma_{\beta\beta}}\right)^{-1} \propto \frac{\Omega^2\tau}{N(H)}\left[1 + \left(\frac{\Delta N \Omega\tau}{N(H)}\right)^2\right]^{-1}.$$

Since $\Omega\tau \approx 1$ at $H = 10^4$ Oe for holes at room temperature, and in strong fields we have $N(H) \widetilde{\propto} H$, it follows that even at $\Delta N/N(0) \approx 10^{-1}$, i.e., for samples that are less pure by a factor ~100 than those employed by us, the influence of the Hall terms is negligibly small and $\rho_{\alpha\alpha} \widetilde{\propto} H$. With decreasing temperature, τ increases, and Kapitza observed, on those samples which were made of less pure material, a deviation from the linear dependence, which was apparently due to the difference between the electron and hole densities.

If, by starting from the foregoing considerations, we determine the change of N as a function of the field in accordance with Kapitza's data[32], and assume that N(60 kOe) coincides with our value, then we obtain $N(270\ kOe)/N(0) \approx 6$, which is in good agreement with the value 5.2 corresponding to results of Brandt et al.[10]

11. It must be emphasized in conclusion that in accordance with our results there exist qualitative contradictions between the experimental and theoretical calculations performed within the framework of the Abrikosov-Fal'kovskiĭ deformation theory when account is taken of four closely-located bands at the points L and T[4,7]. Abrikosov[4] made the first step towards improving the theory by introducing two more bands. According to his results, allowance for a larger number of bands eliminates the previously obtained restriction $\Delta H_{sp}^{-1}/\Delta H^{-1} < 1$ on the spin splitting of the hole levels at $\mathbf{H} \parallel C_3$. Complicating the model further increases the number of parameters describing the carrier spectrum to 12.

At the present time, however, the volume of the experimentally obtained information for bismuth is quite large. Thus, for electrons there are at least 15 parameters. These are: the three dimensions of the Fermi surface, the two values of the curvature at the limiting points, the slope in the basal plane, the parameter describing the asymmetry in the binary plane[16], the three principal values of the effective mass[25], the three principal values of the spin mass, the angle of inclination of the ellipsoid of the spin masses to the basal plane[16,9], and the energy characterizing the threshold of the interband absorption[27]. We should add furthermore that the model should describe the results of magnetooptical investigations[29,30] and the behavior of bismuth in a quantizing field.

The holes are characterized by four quantities: the cross section, mass, and spin splitting at $\mathbf{H} \parallel C_3$ and the cross section at $\mathbf{H} \perp C_3$ ([16,25] and the present paper). Further, the calculated spectrum of the holes should be close to quadratic, and this imposes additional conditions on the relations between the theoretical parameters.

Thus, it seems to us that the large number of phenomenological parameters that appear in the theory is not a principal obstacle to the construction of the theory. It is clear beforehand, however, that the volume of the required work is very large, and to obtain results it is necessary to exert considerable efforts. In our opinion, they can be justified, first, by the fact that the theory describing the spectrum of pure bismuth, at the accuracy which experiment has reached by now, will make it possible to describe from a unified point of view the manifold of phenomena observed in bismuth-antimony alloys. Second, it is very enticing to use the idea of spectrum superposition, on which the Abrikosov-Fal'kovskiĭ theory is based, to construct a theory for such systems as conducting metal oxides. Therefore it seems highly desirable to raise the theory for bismuth to a level such that its validity would become undisputed.

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