Experimental investigation of the "Hall" components of the magnetoresistance tensor of bismuth

Yu. A. Bogod, Vit. B. Krasovitskiĭ, and V. G. Gerasimenko

Physico-technical Institute of Low Temperatures, Ukrainian Academy of Sciences
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The oscillations with period 0.6×10^{-5} Oe^{-1}, observed by the authors in the vicinity of $\mathbf{H} \perp C_3$[4] at hydrogen temperatures, are investigated in the interval 5.6–65 °K in magnetic fields up to 40 kOe. A change in the period is observed for $T > 15$ °K, and the period at 65 °K is 0.47×10^{-5} Oe^{-1}.

Several recent communications report observation of high-frequency quantum oscillations in bismuth. Thus, Brandt, Dolgolenko, and Stupochenko[1] have observed that at $T \approx 0.1$°K and at directions of the magnetic field **H** close to the trigonal axis, the oscillations of the magnetic susceptibility can be represented in the form of a superposition of a fundamental frequency and its multiples. The period of the high-frequency oscillations observed in[1] is close to $(0.5-0.6) \times 10^{-5}$ Oe^{-1}. The period of the de Haas-van Alphen oscillations, namely 0.6×10^{-5} Oe^{-1}, was also observed at 1.3°K and $\mathbf{H} \parallel C_3$ in a later study by Bhargava[2] and was identified with the second harmonic of the electronic oscillations. Brown[3] has reported observation, at helium temperatures and in the vicinity of $\mathbf{H} \parallel C_3$, of a Shubnikov-de Haas oscillation frequency double the value typical of hole Fermi surfaces.

We have observed oscillations of the off-diagonal component of the magnetoresistance with a period 0.6×10^{-5} Oe^{-1}, realized in the vicinity of $\mathbf{H} \parallel C_3$, at hydrogen temperatures[4]. The "high-temperature" oscillations were observed in the interval 5.6–65°K and in magnetic fields up to 40 kOe.

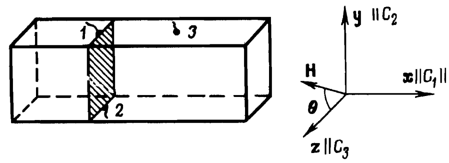

FIG. 1. Geometry of the experiment. 1 to 3—potential contacts (soldered with Wood's alloy). The "Hall" contacts 1 and 2 lie in the plane of rotation of the magnetic field (which is shown shaded).

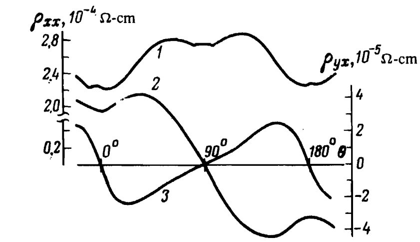

FIG. 2. Rotation diagrams of ρ_{xx} (curve 1) and also of the ρ_{yx} components that are odd (2) and even (3) in the magnetic field; T = 300°K, H = 20 kOe.

RESULTS OF EXPERIMENTS

The measurement procedure was the same as before[5], but in individual cases the electromagnet was replaced by a superconducting solenoid with inside diameter 40 mm. We present below the results obtained mainly with sample Bi-6 with a ratio $R_0^{300}/R_0^{4.2} = 580$, transverse dimensions 6.7×7.2 mm, and distance 5.5 mm between contacts 1 and 3 (Fig. 1). The accuracy of the sample orientation and the degree to which it is a single crystal (we have in mind the relative share of the disoriented blocks) could be assessed from the ρ_{xx} and ρ_{yx} rotation diagrams obtained at room temperature, thereby apparently excluding completely the influence of scattering by the twinning planes, slip planes, and other crystal-structure defects. The sufficient symmetry of the diagrams (see Fig. 2) and the fact that ρ_{yx} satisfies the conditions imposed by the crystal symmetry (identical vanishing at $\mathbf{H} \parallel C_2$ and vanishing of the even component for the case $\mathbf{H} \parallel C_3$) are evidence of the high quality of the sample in the sense indicated above. Notice should be taken of the felicitous position of contacts 1–2, namely at $\mathbf{H} \parallel C_2$ the voltage signal across these contacts, which is proportional to ρ_{xx}, can be noted at helium and hydrogen temperatures only at a sensitivity that exceeds the customary one by three orders of magnitude.

Oscillations with a period $P = 0.6 \times 10^{-5}$ Oe^{-1} were first observed at hydrogen temperatures for values of H from 10 to 20 kOe (Figs. 3 and 4). The monotonic part of $\rho_{yx} \approx \sigma_{yx}/\sigma_{xx}^2$ turn out to be so negligible, that by suitably choosing the sensitivity of the circuit it was possible to record the oscillating signal directly. It must be emphasized that in the entire temperature range the oscillations were registered with one and the same pair of contacts, 1 and 2, and the results obtained in helium agreed well with the data by others, which are gathered in the paper by Bhargava[2] (Fig. 3). Moreover, the oscillations period picked off from these contacts at $\theta = 70°$ and 20°K has the same value as in the helium region, thus also agreeing with the data of[2] (Fig. 4).

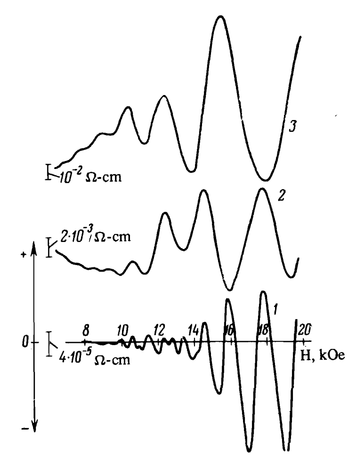

FIG. 3. Oscillations of $\rho_{yx}(H)$ and $\rho_{xx}(H)$, $\theta = 0°$. Curves: 1–$\rho_{yx}(H)$, T = 20°K, P = $0.6 \cdot 10^{-5}$ Oe^{-1}; 2–$\rho_{yx}(H)$, T = 4.2°K, P = $1.23 \cdot 10^{-5}$ Oe^{-1}; 3–$\rho_{xx}(H)$, T = 4.2°K, P = $1.6 \cdot 10^{-5}$ Oe^{-1}.

The strong $\rho_{yx}(\theta)$ dependence (i.e., the large mono-

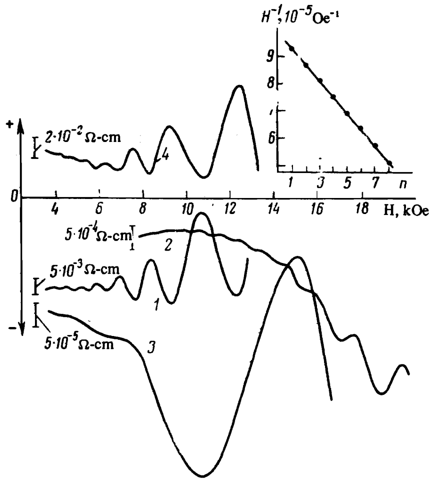

FIG. 4. Oscillations of $\rho_{yx}(H)$ and $\rho_{xx}(H)$. Curves 1, 2—$\rho_{yx}(H)$, $\theta = 10°$, T = 4.2 and 20°K, respectively, P = 2.4 × 10^{-5} Oe^{-1} and 0.6 × 10^{-5} Oe^{-1}. Curve 3—$\rho_{yx}(H)$, $\theta = 70°$K, T = 20°K, P = 6.7 × 10^{-5} Oe^{-1}. Curve 4—$\rho_{xx}(H)$, $\theta = 10°$, T = 4.2°K, P = 2.4 × 10^{-5} Oe^{-1}; n—arbitrarily chosen numbers of the maxima on curve 2.

tonic component of the potential difference transverse with respect to the current), the appearance of the known low-frequency oscillations, and also the change of the projection of the vector of the transverse potential difference on the C_2 direction, all hindered the observation of new oscillations when **H** made angles larger than 30° with C_3. A reliable registration of the extrema with the aid of our procedure could be carried out in the interval ±10° with respect to C_3. At θ equal to zero and ±10°, the period of the observed oscillations differed by not more than 10—20% from the estimates in the interval $-30° < \theta < +30°$, and amounted to 0.6 × 10^{-5} Oe^{-1}. (The period of the oscillations was determined from the maxima of ρ_{yx} (see Fig. 4). It could be determined just as successfully from the minima, inasmuch as both families of the extrema yield the same value of the period P.)

As shown by an analysis of the curves, the decisive contribution to the oscillations of the "Hall" component (including the case $\theta = 0°$) is made by the even component. On the other hand, we have already mentioned that in a magnetic field parallel to the trigonal axis the terms even in the magnetic field in the off-diagonal components of the magneto-resistance tensor should be equal to zero. But since the direction H ∥ C_3 is a mathematical point on the rotation diagram, which is practically impossible to hit, and furthermore, the orientation of the crystal is inevitably subject to a certain error, the presence of a component even in the magnetic field for the direction $\theta = 0°$ (which strictly speaking does not coincide with H ∥ C_3) does not contradict symmetry.

Figure 5 shows the oscillations of ρ_{yx} at $\theta = 0°$, recorded in the temperature interval 5—65°K and in magnetic fields up to 40 kOe. It is clearly seen that oscillations with a different frequency, whose relative share increases with increasing temperature, appear against the background of the long-period oscillations that are characteristic of the helium region[1]. At T = 10°K, the "high-temperature" oscillations predominate completely.

At first glance, the picture of the high-temperature oscillations for H > 20 kOe becomes more complicated,

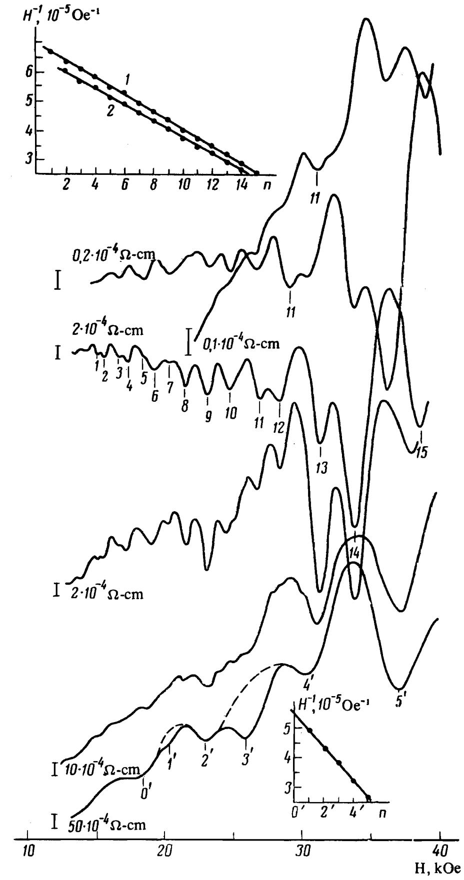

FIG. 5. Oscillations of $\rho_{yx}(H)$, $\theta = 0°$. The curves correspond to the following temperatures (reading up): 5.6, 8, 10, 14.5, 45, and 65°K. 2, 4, 6, . . . , and 1, 3, 5, . . . mark families of equivalent minima. Dashed curve—oscillations without allowance for the spin splitting at T = 5.6°K. Curve 1 (in upper insert) corresponds to T = 14.5°K, P = (0.6 ± 0.02) × 10^{-5} Oe^{-1}; curve 2—T = 45°K, P = 0.54 ± 0.02) × 10^{-5} Oe^{-1}. The curve on the lower insert was plotted for T = 5.6°K and P = (1.12 ± 0.02) × 10^{-5} Oe^{-1}.

but at a fixed temperature and when plotted in the coordinates H^{-1} and n, where n are arbitrarily chosen integers, all the minima fit one straight line (see the insert of Fig. 5). The oscillation period determined from the equivalent minima on the curves remains practically unchanged to 20°K and is equal to 0.6 × 10^{-5} Oe^{-1}. (The fact that a single straight line in the (H^{-1}, n) plane fits the equivalent and nonequivalent points is a consequence of the equality of the distances between all the minima of the oscillating part of ρ_{yx} when plotted against the reciprocal of the field.) Further lowering of the temperature is accompanied by a change in the period. Thus, for example, T = 45°K corresponds to P = 0.54 × 10^{05} Oe^{-1}, and T = 65°K, corresponds to P = 0.47 × 10^{-5} Oe^{-1} (see Fig. 6). If we compare the picture of the oscillations at 14 and 45°K, a noticeable difference is observed in the growth increment of the amplitude with increasing magnetic field, in that the higher temperature corresponds to a larger increment. As seen from Fig. 5, in a given magnetic field the amplitude of the new oscillations decreases with increasing temperature, and in the region where the oscillation period remains practically unchanged (8—20°K), the damping of the amplitude is best described by the relation

$$\tilde{\rho}_{yx} \sim Te^{-\alpha T}, \qquad (1)$$

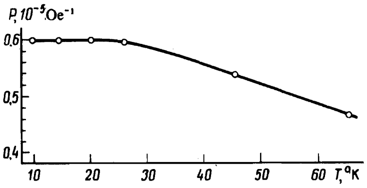

FIG. 6. Temperature dependence of the new period of the oscillations.

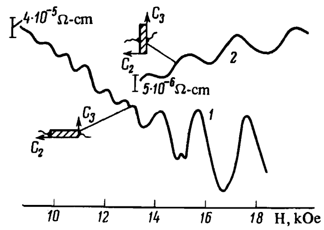

FIG. 7. Oscillations of $\rho_{yx}(H)$, T = 20°K. Curves 1 and 2 correspond to samples Bi-8 (cross section 6 × 0.6 mm, H ∥ C_3, P = 0.65 × 10^{-5} Oe^{-1}) and Bi-7 (0.8 × 6.5 mm, the angle between H and C_3 is 8°, P = 0.7 × 10^{-5} Oe^{-1}). The positions of the "Hall" contacts are shown in the figure.

where $\alpha = 0.15$ at H = 19 kOe. The right-hand side of (1) can be regarded (apart from a constant factor) as the asymptotic expression for the first term of the series

$$\sum_{r=1}^{\infty}\frac{(-1)^r}{r^{1/2}}\frac{\alpha rT}{\operatorname{sh}(\alpha rT)} \qquad (2)$$

which is valid at $\alpha T > 1$. To extrapolate $\tilde{\rho}_{yk}$ in the region T < 8°K, we therefore used relation (2), which yields at T > 8°K the same values of $\tilde{\rho}_{yx}(T_1)/\tilde{\rho}_{yx}(T_2)$, as (1). Thus, at T = 4.2°K, we obtain $\tilde{\rho}_{yx} = 6 \times 10^{-4}$ Ω-cm, i.e., a value smaller by a factor 17 than the fundamental harmonic of the known oscillations.

The high-temperature oscillations of ρ_{yx} are reproducible from sample to sample. This is illustrated in Fig. 7, which shows the oscillations of ρ_{yx} recorded at 20°K in the region H ∥ C_3, for the single crystals Bi-7 and Bi-8 with a ratio $R_0^{300}/R_0^{4.2}$ equal to 260 and 475, respectively. At 20°K we have observed also high-temperature oscillations of ρ_{xx}, the amplitude of which in a magnetic field H = 30 kOe was larger by one order of magnitude than the amplitude for ρ_{yx}. The value of $\tilde{\rho}_{xx}/\rho_{xx}$ at hydrogen temperatures is, however, ≈1%, so that from the experimental point of view the study of the high-temperature oscillations of ρ_{xx} is much more difficult than for ρ_{yx}.

DISCUSSION OF RESULTS

It should be noted first that the high-temperature oscillations of the magnetoresistance of bismuth have all the characteristic attributes of Shubnikov-de Haas oscillations: they are periodic in the reciprocal field, they attenuate with increasing temperature at a given value of the magnetic field, and the higher the temperature the faster the increase of the amplitude with increasing magnetic field (Fig. 5). Further, a decrease in the period of the oscillations for T > 15°K offers evidence of the temperature growth of the carrier density in bismuth. Indeed, the deformation potentials for bismuth are equal approximately to 10 eV, and the change of the lattice anisotropy $\Delta(c/a)$ when the sample is cooled from 300 to 4.2°K is 0.2°[6]. Assuming that an increase of the temperature from 4 to 70°K leads to $\Delta(c/a) \sim 10^{-3}$, we obtain for the change of the Fermi level $\Delta\epsilon_F \sim 10$ meV, which corresponds precisely to 30–50% of the change in the concentration observed in a number of studies (see, e.g.,[7]).

According to Adams and Holstein[8], for finite temperatures and in the case $\hbar\omega_c < \epsilon_F$ (ω_c is the cyclotron frequency and ϵ_F is the Fermi energy), the amplitude of the oscillations of the magnetic conductivity is expressed in the form of the series

$$\tilde{\sigma} \sim \frac{Ne^2}{m^*\omega_c^2\tau}\frac{5}{2}\left(\frac{\hbar\omega_c}{\epsilon_F}\right)^{1/2}\sum_{r=1}^{\infty}\frac{(-1)^r}{r^{1/2}}\frac{2\pi^2krT/\hbar\omega_c}{\operatorname{sh}(2\pi^2krT/\hbar\omega_c)}. \qquad (3)$$

Ignoring the absence of the fundamental harmonic at T > 10°K, we assume that the high-temperature oscillations, if they are of high frequency, are due to the next higher terms of (3), and we estimate the damping of the amplitude as the temperature is increased from 10 to 20°K. For the known cyclotron masses in the H ∥ C_3 direction[9] we have $2\pi^2kT/\hbar\omega_c > 1$ and therefore, replacing the sine function by the hyperbolic exponential and assuming for simplicity r = 2, we get at H = 20 kOe, disregarding the temperature dependence of the relaxation time,

$$\tilde{\rho}_{yx}(10°\,K)/\tilde{\rho}_{yx}(20°\,K) \sim 5\cdot 10^6,$$

whereas the experimental results yield a value on the order of unity. It is therefore perfectly clear that the observed oscillations cannot be interpreted with the aid of higher harmonics within the framework of the existing theory.

We now present arguments from which it follows that the period of the high-frequency oscillations (naturally, again within the framework of the known concepts) is not connected with the partial disorientation of the crystal:

1. The disoriented regions (including the twin layers) in the Bi-6 sample occupy a value much smaller than the main matrix. This follows from the symmetry of the rotation diagrams of the magneto-resistance-tensor components (Fig. 2).

2. The observed period is well reproducible both in experiments with a single crystal and on going from sample to sample.

3. Periods close to 0.6×10^{-5} Oe^{-1} in bismuth correspond to cyclotron masses $m^* \sim 0.1m_0$. The amplitude of the oscillations connected with such a cyclotron mass should attenuate by more than 10^6 times when the temperature is increased from 4 to 20°K in a magnetic field H = 20 kOe, and by more than 10^4 times when the temperature is increased from 10 to 20°K. Actually, as already mentioned, when the temperature changes from 10 to 20°K the damping of the amplitude is of the order of unity. In addition, by measuring $\tilde{\rho}_{yx}$ for the case T = 20°K and H = 20 kOe and estimating the amplitude expected in the helium region, we obtain a value that exceeds by several orders of magnitude the monotonic part of ρ_{xx} at 4.2°K, which is impossible.

4. The damping of the oscillation amplitude with increasing temperature in the interval 8–20°K, where the period remains practically unchanged, is adequately described by relation (3). The cyclotron mass determined from the temperature dependence of the amplitude (see formulas (1) and (2), in which one must put $\alpha = 2\pi^2k/\hbar\omega_c$) is $0.016m_0$. According to the universally accepted model of the electron spectrum of bismuth, this cyclotron mass corresponds to a period which is larger than the observed one by approximately one order of magnitude. In addition, at $m^* = 0.016\,m_0$ and $\epsilon_F \approx 25$ meV, the condition $\hbar\omega_c \gtrsim \epsilon_F$ is satisfied in magnetic fields 20–25 kOe, so that the oscillations connected with the given group of carriers should vanish.

Yet near 40 kOe, in the indicated temperature interval, a minimum is observed and the extrema as functions of the reciprocal field remains equidistant up to this minimum.

The constancy of the period of the known oscillations in the interval 4–20°K, observed when the direction of H is close to that of C_3 (see Fig. 4), and also the character of the curves in Fig. 5, contradict the assumption that a phase transition is realized in bismuth and can be the cause of the appearance of the new period of oscillations.

For an unambiguous interpretation of the observed phenomenon, detailed investigations are being carried out of the angular dependence of the period of the oscillations in different crystallographic planes.

[1)]The difference between the periods in the helium region on the plots of Figs. 3 and 5 is apparently due to a small change in the orientation during the remounting of the sample, which was placed in a different cryostat for the measurements in a wider range of temperatures and magnetic fields.

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