Quantum size oscillations of the magnetoresistance of thin-filament bismuth single crystals

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The longitudinal magnetoresistance of cylindrical single crystals (filaments) of bismuth $0.1 \leqslant d \leqslant 20\ \mu$m in diameter has been studied in magnetic fields up to 50 kOe at 4.2 K. The filaments with $d \leqslant 1\ \mu$m exhibit two types of quantum size oscillations in the magnetoresistance; these two types of oscillations differ in physical nature. Classified as oscillations of the first type are two groups of oscillations which stem from the discrete energy spectrum of the "bulk" electrons and holes. In fields $0 < H \leqslant H^{e}_{\text{cut}}$, where H^{e}_{cut} is the electron cutoff field, the predominant oscillations are those of the first group which stem from electrons and whose periods in units of the magnetic flux, $\Delta\Phi = (1/4)\Delta H p d^2$, lie in the interval $(1-3)\Phi_0$, where Φ_0 is the quantum of flux in the normal metal. In fields $0 < H \leqslant H^{h}_{\text{cut}}$, where H^{h}_{cut} is the cutoff field for holes, high-frequency quantum size oscillations of the Dingle type (the second group) are observed, with periods in units of the flux, $\Delta\Phi \approx (0.3-0.9)\Phi_0$, which correspond to the hole part of the Fermi surface of bismuth. Classified as oscillations of the second type are some quantum size oscillations of the longitudinal magnetoresistance with periods $\Delta\Phi \approx (2–26)\Phi_0$, which are observed for the first time in this study. These oscillations are seen at fields well above the fields H_{cut} set by electrons and holes. The oscillations of this type may be due to carriers which are tracing out "jumping" trajectories along the surfaces of the filaments.

1. INTRODUCTION

Size effects can be classified as either semiclassical effects, in which one parameter of the trajectory of an electron becomes comparable to the dimension of the sample, or quantum effects, in which the energy of the motion of the electrons is quantized in a sample of bounded size. Semiclassical size effects have been studied in several places, but there have been relatively few studies of quantum size effects. The reason for this situation is that the samples which would be required for an observation of quantum size effects would have dimensions comparable to the de Broglie wavelength of the charge carriers. Quantum size effects were originally observed in a study of epitaxial films of bismuth[1] which consisted of a mosaic of crystallites which had longitudinal dimensions significantly greater than their thickness and which were oriented in a definite way. Since the thickness of the crystallites must be comparable to the wavelength of the electrons if quantum size effects are to be observed, some very stringent requirements are imposed on the homogeneity of the epitaxial films over thickness. Some other objects in which quantum size effects can be observed are lamellar crystals and filamentary whiskers. The complexity of working with such objects, however, has kept the list of such studies very short.[2,3]

Some objects which are very interesting for studies of quantum size effects but which have received little study in practice are thin cylindrical filamentary crystals in glass insulation,[4,5] synthesized by the Ulitovskiĭ method.[6] These crystals have several advantages over epitaxial films and whiskers. The most important of these advantages are that it is possible to synthesize single-crystal samples with a thickness which varies monotonically without a variation in crystallographic orientation; the cylindrical shape is very regular; they are highly uniform in the transverse direction; it is possible to reproduce samples with given dimensions and orientations; the quality of the surface is high; the surface is protected from oxidation; the samples are convenient to work with because of the glass jacket protecting the crystalline core; and the geometry is convenient for theoretical work.

Quantum size effects in cylindrical filamentary crystals were first studied in Refs. 4 and 5. Quantum-size oscillations in the magnetoresistance with two characteristic periods $(\Delta H \sim d^{-2})$ as functions of the magnetic flux, $\Delta\Phi = \Delta H \pi R^2$ (R is the radius of the cylinder), were observed in bismuth filaments $0.2 \leqslant d \leqslant 0.8\ \mu$m in diameter in a longitudinal magnetic field $\mathbf{H} \| \mathbf{j}$ (j is the current density). These periods were $\Delta\Phi_1 = hc/e = \Phi_0$ (h is Planck's constant, c is the velocity of light, e is the charge of an electron, and Φ_0 is the quantum of flux in a normal metal) and $\Delta\Phi_2 = 1.4\Phi_0$. The quantum size oscillations took the form of beats and were observed in magnetic fields in which the Larmor radius of the electrons satisfies $r_H > R$. As the magnetic field was increased, the amplitude of the oscillations decreased, vanishing by the time at which Shubnikov-de Haas oscillations due to electrons appeared.

In the present paper we report a study of the longitudinal magnetoresistance of bismuth filaments with $0.1 \leqslant d \leqslant 20\ \mu$m at liquid-helium temperatures and also a systematic experimental study of quantum size oscillations of cylindrical single crystals of thickness $d \leqslant 1\ \mu$m. This study was carried out in order to obtain information in as much detail as possible on the particular features of the quantum size effect over a broad range of magnetic fields.

2. THEORY

Quantum size oscillations in thermodynamic properties of thin cylindrical conductors in a weak magnetic field

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$$H \ll H_{cut} = cp_F/eR, \quad (1)$$

where H_{cut} is the cutoff magnetic field, at which the diameter of the electron orbit is equal to the cylinder diameter $2R$, and p_F is the Fermi momentum, were first studied by Dingle.[7] It follows from Dingle's calculations that in fields satisfying (1) there are a multitude of frequencies, which are spaced equidistantly as a function of H. This problem for thin single-crystal films were solved by Lifshitz and Kosevich.[8] In weak magnetic fields, $H < H_{cut}$, where Landau magnetic quantization is unimportant, the oscillations in the magnetoresistance stem from size quantization of the electron energy spectrum. As the magnetic field or the filament thickness is varied, the discrete energy levels cross E_F (the Fermi energy of the charge carriers) and thereby cause oscillations in the state density at the Fermi boundary.

The behavior of thin cylindrical conductors in weak magnetic fields $r_H \gg R$ was studied theoretically in Refs. 9–11. It was shown there that the discrete electron spectrum which stems from the size quantization gives rise to two terms, differing in period and amplitude, in the expression for the state density. One term, with the larger amplitude and a complex, nonmonotonic dependence on the magnetic flux $\Delta\Phi = \Delta HS = \Delta H\pi R^2$, resulted from electrons whose trajectories pass near the axis of the cylinder ("bulk" electrons). These electrons are responsible for the quantum size oscillations of the Dingle type. The second term, with an amplitude smaller by a factor of $(k_F R)^{1/2}$ (k_F is the Fermi wave vector of an electron), varies periodically as a function of the flux with a universal period $\Delta\Phi = \Phi_0 = hc/e$. This term corresponds to "grazing" electrons, which are localized in a narrow layer near the surface. The spectrum of these levels is quite different from that of the magnetic surface levels at a plane boundary.[12] Incorporating the diffuse nature of the scattering of carriers at the surface makes it possible to effectively distinguish between the contributions of the different electron groups. Bogachek *et al.*[9–11] showed that the angular dependence of the coefficient of specular reflection keeps the amplitude of the Dingle oscillations small under ordinary conditions, so that the oscillations in the thermodynamic potential are dominated by the grazing electrons, whose spectrum is spread out less than that of the bulk electrons. The longitudinal kinetic coefficients of cylindrical conductors thus vary periodically as the flux Φ is varied, with a period equal to the quantum of flux in a normal metal, hc/e. This quantum does not depend on the particular nature of the dispersion law for the charge carriers.

To refine the interpretation of the quantum size oscillations discovered in Refs. 4 and 5, Mankin[13] and Nedorezov[14] calculated kinetic and thermodynamic characteristics of a size-quantized wire with reference to thin bismuth filaments. Using Dingle's theory,[7] Mankin calculated the oscillatory part of the conductivity, σ_{osc}, at weak magnetic fields under the condition[13] $r_H \gg R$. The dispersion law for electrons in the plane perpendicular to the axis of the wire was assumed to be isotropic. The expression which he derived for σ_{osc} contains a sum of a large number of oscillatory components, some of which oscillate in a parallel field H with a period ΔH which is inversely proportional to the cross-sectional area of the wire, S:

$$\Delta_{ML} H = \frac{2\pi\Phi_0}{SM\sin(2\pi L/M)}, \quad (2)$$

where $M = 3, 4, 5, \ldots$; and $L = 1, 2, 3, \ldots, M/2$. According to (2), the periods of the first nine oscillatory components, in terms of the dimensionless units $\Delta HS/\Phi_0$, are 2.36, 1.57, 1.32, 2.14, 1.21, 1.17, 0.92, 1.45, and 1.13. Essentially all of the frequencies except the first, second, and fourth fall in the frequency interval observed in Refs. 4 and 5.

Nedorezov[14] generalized Dingle's theory to the case of conduction electrons with an anisotropic quadratic dispersion law. He calculated the quantum energy levels of conduction electrons for the case of specular reflection from the boundary of a cylindrical sample and derived expressions for the state density and thermodynamic properties in weak magnetic fields, $H \ll H_{cut}$. The oscillatory part of the thermodynamic potential which was calculated in Ref. 15 contains a sum of oscillatory components which are equidistant along the magnetic-field scale. As an application of the theory, Nedorezov[14] analyzed the oscillations in the magnetic susceptibility of electrons in a bismuth single-crystal cylinder with axis running along the ΓL direction in the bisector-trigonal plane of the Brillouin zone. For a crystallographic orientation of this type, which is characteristic of the bismuth microcylinders studied in Refs. 4 and 5, the direction in which one of the electron ellipsoids (the a ellipsoid) is stretched out makes a small angle ($\sim 20°$) with the axis of the filament. The area of the intersection of this ellipsoid with a plane perpendicular to the axis of the filament is close to a minimum (see the inset in Fig. 1). The other two electron ellipsoids (b ellipsoids) are positioned symmetrically with respect to the axis of the cylindrical single crystal, and their intersection with the plane perpendicular to the axis of the filament is $S_b \approx 2S_a$. It follows from the estimates found in Ref. 14 that the terms referring to the a ellipsoid should dominate the quantum-size oscillations of the magnetic susceptibility of cylindrical bismuth single crystals. For this ellipsoid, the periods of the five most important oscillatory components, in units of $\Delta HS/\Phi_0$, are 2.60, 1.61, 1.35, 1.22, and 1.15.

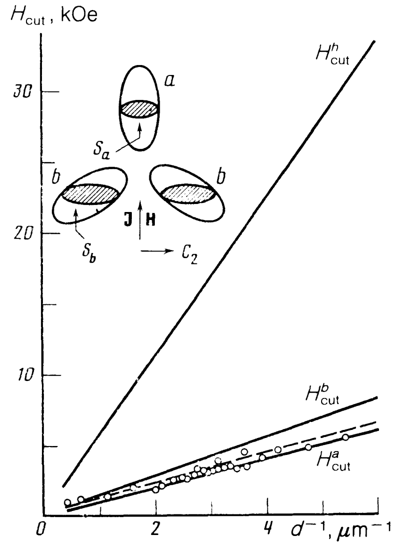

FIG. 1. Calculated cutoff fields for the electron (H^a_{cut}, H^b_{cut}) and hole (H^h_{cut}) Shubnikov–de Haas oscillations and experimental determinations of H_{max} versus d (dashed line) for the Bi single crystals studied. Here H_{max} is the field at which $\rho(H)$ goes through its maximum. The inset shows the arrangement of the electron constant-energy surfaces with respect to the longitudinal axis of the filament, which coincides with the direction of the magnetic field.

In addition to the mechanisms discussed above, there is yet another, which leads to a quantization of the energy of electrons in bounded conductors. Near a plane metal surface with specular reflection, in a magnetic field parallel to the surface, some of the electrons move along cyclic ("jumping") trajectories. Their energy is quantized. As a result, a system of energy levels called "magnetic surface levels" appears. Oscillations in the surface impedance which are associated with these levels and which arise from a resonant absorption of microwave photons in very weak fields (~ 1 Oe) were first observed in bulk bismuth single crystals in Ref. 12. It was also suggested that the magnetic surface levels in filamentary or film single crystals of thickness $d \sim Z_z$, where Z_n is the height of the arc of the truncated orbit of the electrons, can lead (in particular) to Shubnikov–de Haas-type oscillations in the magnetoresistance.

It thus follows from an analysis of the theoretical papers that quantum size oscillations of the following types may be observed in size-quantized cylindrical single crystals with a specular-reflection surface: 1) quantum size oscillations of the Dingle type,[7] which stem from charge carriers which collide with the surface at large angles ("bulk" carriers) in magnetic fields $H < H_{\rm cut}$; 2) quantum size oscillations of the flux quantization type[9–11] ($H \ll H_{\rm cut}$), which are due to electrons which have large values of the magnetic quantum number m, are localized in a narrow layer near the surface, and which move along closed trajectories; and 3) oscillations of the magnetoresistance which arise because magnetic surface levels are produced in a magnetic field by charge carriers which are "jumping" along the surface.[12]

3. TEST SAMPLES AND MEASUREMENT PROCEDURE

Thin single-crystal filaments of bismuth were synthesized by the Ulitovskiĭ method.[6] The thickness of the glass insulation on the samples, whose diameter was $d \leqslant 1\ \mu$m, was $\sim 5\ \mu$m. Laue patterns verified that the filaments were single crystals; their orientation was determined from rotation x-ray diffraction patterns. Essentially all of the filaments studied had the following orientation. The bisector axis C_1 made an angle of 20° with the filament axis, while the trigonal axis C_3 made an angle of 70° with it. One of the binary axes C_2 ran perpendicular to the axis of the cylinder. The diameter of the filaments was measured with an MIN-8 microscope (1350x) within an error of 0.1 μm. The precise diameter of the filamentary crystals of thickness $d \leqslant 1.5\ \mu$m was determined by calculations based on the resistance at a temperature of 300 K. The length of the filamentary samples was 2–4 μm and was measured within an error of less than 10 μm by an MI-1 microscope. The samples were mounted on substrates of a foil-coated glass-cloth-base laminate. The current leads and potential leads were attached with gallium solder. The longitudinal magnetoresistance was measured in the field of a superconducting solenoid ($H \leqslant 50$ kOe) at liquid-helium temperature in an apparatus which automatically recorded curves of $\rho(H)$, $\partial\rho(H)/\partial H$ and $\partial^2\rho(H)/\partial H^2$. During the recording of the magnetic-field dependence of the first and second derivatives of the magnetoresistance with respect to the magnetic field, we used a standard modulation technique. A study of the second derivative of the resistance with respect to the magnetic field made it possible to minimize the effect of both the monotonic change in the magnetoresistance and the Shubnikov–de Haas oscillations on the quantum size oscillations. Minimizing these effects is particularly important in cases in which the quantum size oscillations are observed against the background of Shubnikov–de Haas oscillations. The magnitude of the measurement current flowing through the filamentary single crystals was in the Ohm's-law region.

Since the quantum size oscillations consist of a superposition of many frequencies, we carried out a numerical Fourier analysis of the oscillation curve to determine the periods of these oscillations. The procedure which we used is based on the results of Ref. 16, where a filter with a Gaussian distribution was used to suppress harmonic frequencies. The values found for the frequencies and amplitudes were refined by the method of least squares based on five points. This procedure improved the accuracy of the determination of the periods by an order of magnitude. To improve the resolution of the constituent frequencies, we recorded the oscillation curves to be subjected to the Fourier analysis in an expanded scale along H. This approach also made it possible to obtain enough experimental point for an accurate determination of the periods. Since the high-frequency quantum size oscillations were observed over a broad range of the magnetic field, the oscillation curves were recorded part by part on separate sheets and then joined. The Shubnikov–de Haas oscillations were subtracted from the oscillation pattern.

4. EXPERIMENTAL RESULTS

1. Longitudinal magnetoresistance and Shubnikov–de Haas oscillations of filamentary bismuth single crystals

The longitudinal magnetoresistance of bismuth filaments with $d > 12\ \mu$m exhibits the same features as bulk samples of the corresponding crystallographic orientation: In fields up to ~ 1 kOe, one observed a sharp increase in the magnetoresistance. This increase is followed by a broad magnetic-field region in which the resistance increases more slowly. The longitudinal magnetoresistance reaches a maximum at $H = 30 \pm 4$ kOe. A further increase in the magnetic field results in a decrease in the resistance; the resistance continues to decrease up to the strongest fields.

As the diameter of the bismuth filaments is reduced, the nature of the $\rho(H)$ curves changes substantially. Figure 2 shows a typical magnetic-field dependence of the longitudinal magnetoresistance at 4.2 K in fields up to 50 kOe for bismuth single-crystal filaments with $d < 4\ \mu$m. In weak magnetic fields, there is a very slight increase in the resistance, up to a field $H_{\rm max}$, whose value increases with decreasing filament diameter. A further increase in the magnetic field has the result that the resistance of the filamentary samples decreases, reaching a level of 25–40% of its values in a zero field at ~ 50 kOe. Studies of the longitudinal magnetoresistance of a large number of filaments of various diameters showed that for samples of thickness $d < 2\ \mu$m we have $H_{\rm max} \propto d^{-1}$, while the product $H_{\rm max} d$ is 1.1 μm·kOe (Fig. 1). In fields $H \geqslant H_{\rm max}$, Shubnikov–de Haas oscillations appear on the magnetic-field dependence of the longitudinal magnetoresistance; these oscillations are clearly visible against the background monotonic variation in $\rho(H)$ (Fig. 2). To bring out the oscillatory part of the magnetoresistance better, we recorded its derivative with respect to the magnetic field. The curve of $\partial\rho(H)/\partial H$ in Fig. 2 is dominated by Shubnikov–de Haas oscillations with a period

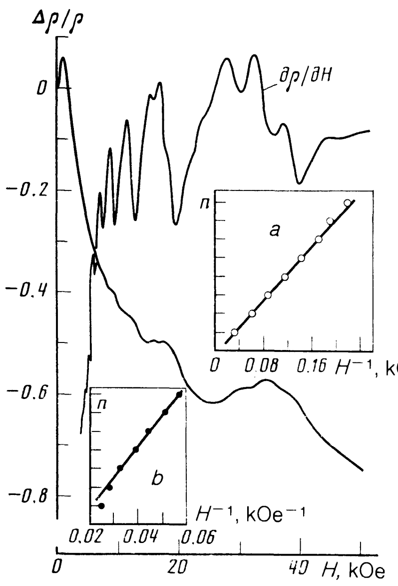

FIG. 2. Longitudinal magnetoresistance and its derivative (arbitrary units) with respect to the magnetic field for a Bi sample with $d = 1.8\,\mu$m at 4.2 K. The inset show the nominal quantum index of the Shubnikov–de Haas oscillations versus the reciprocal of the magnetic field for (a) the electrons of b ellipsoids and (b) the holes.

$\Delta(H^{-1}) = 2.8\cdot 10^{-5}$ Oe^{-1}, which correspond to the electrons of the b ellipsoids which are positioned symmetrically with respect to the axis of the filament (see the inset in Fig. 1). The maximum on the $\rho(H)$ curve at $H\,\|\,36$ kOe corresponds to the emergence of the last Landau levels (the 0^+1^- doublet) from this group of electrons. In fields $H\geqslant 10$ kOe, Shubnikov–de Haas oscillations due to holes become superimposed on the oscillations due to electrons. The period of the hole oscillations determined in weak fields is $\Delta(H^{-1}) = 0.62\cdot 10^{-5}$ Oe^{-1}. The Shubnikov–de Haas oscillations from the a ellipsoid electrons, with a period $\Delta(H^{-1}) = (7.7\pm 0.5)\cdot 10^{-5}$ Oe^{-1}, are detected primarily in filaments with $d > 4\,\mu$m. The Shubnikov–de Haas oscillations from the b-ellipsoid electrons are observed in samples of all diameters. The period of these oscillations, $\Delta(H^{-1}) = (3.3\pm 0.5)\cdot 10^{-5}$ Oe^{-1}, appears to be independent of the thickness. Figure 1 shows calculated cutoff fields for the electrons of the a and b ellipsoids and for the holes; beginning at these cutoff fields, the conditions ($r_H \leqslant R$) for observation of the Shubnikov–de Haas oscillations from the corresponding group of charge carriers become satisfied in the filamentary bismuth single crystals. Also shown here is an experimental curve of $H_{\rm max}$ versus d for the filamentary single crystals studied in the present experiments.

The presence of a negative magnetoresistance (Fig. 2) and the adherence to the behavior $H_{\rm max} \propto d^{-1}$ (Fig. 1) are evidence that a galvanomagnetic size effect is occurring in the thin bismuth filaments[15,17]: Some of the electrons (or holes) gyrate in the magnetic field and avoid meeting the surface. The effect is to "improve" the conductivity of the filament. In the filamentary bismuth single crystals the galvanomagnetic size effect is manifested against the background of the "bulk" magnetoresistance which is due to the anisotropy of the Fermi surface and which determines the increase in the resistance in fields $0 < H < H_{\rm max}$. At $H > H_{\rm max}$ the decrease in the resistance due to the galvanomagnetic size effect outweighs the increase in the "bulk" part of $\rho(H)$. Significantly, the large magnitude of the negative magnetoresistance of the filamentary bismuth single crystals with $d < 4\,\mu$m in strong magnetic fields results from the fact that at $H > H^b_{ql}$ (H^b_{ql} is the quantum-limit field for the electrons of the b ellipsoids) the electrical conductivity also begins to increase as a result of an increase in the density due to the flow of electrons out of the valence band into the conduction band.[18] It is this effect which leads to the decrease in the resistance of the large-diameter filaments for $H > 30$ kOe. Since the Shubnikov–de Haas oscillations due to the electrons begin at fields $H \approx H_{\rm max}$, we can make use of the $H_{\rm max} \propto d^{-1}$ dependence to determine the extreme value of the transverse Fermi momentum of the electrons. According to (1), the experimental value $H_{\rm max} d = 1.1\,\mu$m·kOe (Fig. 1) corresponds to $p_F^{\rm extrm} = 0.88\cdot 10^{-21}$ g·cm/s. Using the known values of the parameters of the Fermi surface of bismuth[19] and the crystallographic orientation of the filaments, we find that the values of $p_F^{\rm extrm}$ in the plane perpendicular to the axis of the filament are $0.80\cdot 10^{-21}$ and $1.10\cdot 10^{-21}$ g·cm/s for the electrons of the a and b ellipsoids, respectively, and $4.46\cdot 10^{-21}$ g·cm/s for the holes. The value found experimentally for $p_F^{\rm extrm}$ for the bismuth filaments should probably be regarded as an average value over all of the electron ellipsoids.

The presence of Shubnikov–de Haas oscillations from both the electron and hole parts of the Fermi surface indicates that these filamentary bismuth crystals are of high quality. The periods of the Shubnikov–de Haas oscillations from L electrons and T holes found for them agree well with the corresponding periods which are characteristic of bulk samples. It was found that the period of the Shubnikov–de Haas oscillations does not change down to a thickness $d = 0.1\,\mu$m; this result is evidence that the concentration of charge carriers remains constant in filamentary bismuth single crystals with $d\geqslant 0.1\,\mu$m.

2. Quantum size oscillations in the longitudinal magnetoresistance

The general picture of the quantum size oscillations observed in cylindrical single-crystal filaments of bismuth is unusually complicated. However, a study of a large number of samples (more than 50 in the present experiments) varying in diameter, surface quality, and volume makes it possible to distinguish in the overall frequency spectrum various groups of oscillations differing in periods, in the magnetic-field intervals in which they are seen most clearly, and in the dependence of the amplitude on the magnetic field, although the different types of oscillations are usually superimposed on each other in the highest-quality samples.

For all the samples, we observe size oscillations on the curve of $\rho_\|(H)$ in the initial interval of magnetic fields, where the condition $H < H_{\rm cut}$ holds; the parameter values of these oscillations correspond to the oscillations which were observed in Refs. 4 and 5. The amplitude of such oscillations on the $\rho_\|(H)$ curve is usually at a maximum. However, differentiating twice reveals new groups of size oscillations over a broad range of magnetic fields. The parameters of these new groups of oscillations are quite different from those of the "classical" oscillations which were observed in Refs. 4 and 5. The following classification includes both the classi-

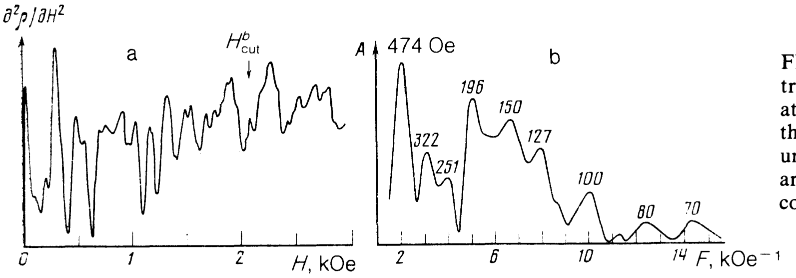

FIG. 3. a—Quantum-size oscillations; b—their Fourier spectrum, for a filamentary Bi sample with a diameter $d = 0.68\ \mu m$ at $T = 4.2$ K. Shown on the spectrogram is the dependence of the amplitude (A) of the quantum size oscillations, in arbitrary units, on the frequency F, in reciprocal kiloersteds. The peaks are labeled with the absolute value of the period of the oscillation component, ΔH, in oersteds.

cal oscillations and the oscillations of the new types.

In the first group we can place the quantum size oscillations with periods of $(1\text{–}4.2)\Phi_0$, which are seen in the initial region of magnetic fields. The particular features of these oscillations are shown quite clearly in Fig. 3, which is a plot of the quantum size oscillations in the longitudinal magnetoresistance of a filamentary bismuth sample $0.68\ \mu m$ thick. The oscillations are of the nature of clearly defined beats, and their period ΔH is constant in a "forward" magnetic field and is inversely proportional to the cross-sectional area of the filament. With increasing H, the amplitude of the quantum size oscillations decreases, but the damping rate in a magnetic field differs for the different frequencies. By the time the Shubnikov–de Haas appear in fields $H \approx H_{max}$, the quantum size oscillations have been completely damped in most of the filaments with thicknesses $d \geqslant 0.3\ \mu m$. However, there are some filamentary single crystals in which quantum size oscillations are detected even at fields above H_{max}.

In the second group of oscillations are the high-frequency quantum size oscillations with periods of $(0.3\text{–}0.9)\Phi_0$ which are observed over a significantly broader range of magnetic fields. These oscillations are seen particularly clearly in bismuth samples with $d = 0.40\ \mu m$ (Fig. 4). In fields $H > H_{max} = 3.4$ kOe, high-frequency quantum size oscillations are superimposed on electron Shubnikov–de Haas oscillations which arise. Although the amplitude of these oscillations is small in comparison with the amplitude of the Shubnikov–de Haas oscillations, they can be reliably detected on the curves of $\partial\rho/\partial H$ at fields up to $H \approx 14$ kOe, and they are completely reproducible when the measurements are repeated. The lower curve in Fig. 4 shows that when we record the second derivative of the magnetoresistance with respect to the magnetic field we can bring out the size-dependent oscillatory part of $\rho(H)$ more clearly. In this case, however, the actual relation between the amplitudes of the low- and high-frequency components is skewed in favor of the latter, by a factor of about 2.5. This circumstance must always be taken into consideration in discussing the contribution of some frequency or other to the oscillatory dependence.

The quantum size oscillations of this group are seen most frequently in filamentary samples with $d < 0.3\ \mu m$. Figures 5–7 show experimental curves found for filaments with thicknesses in this region. A common feature for all three samples is the predominance in weak magnetic fields, $H < H_{max}$, of the quantum size oscillations of the first group, with periods $\Delta\Phi \sim (1 - 4.2)\Phi_0$ in the flux. In fields $H > H_{max}$, superimposed on the Shubnikov–de Haas oscillations, we can clearly see high-frequency quantum size oscillations with periods $\Delta\Phi \sim (0.3 - 0.9)\Phi_0$. The magnetic-field interval over which these oscillations exist expands with decreasing filament thickness. The picture of the superposition of the high-frequency quantum size oscillations on the Shubnikov–de Haas oscillations can be seen particularly clearly in the samples with $d \sim 0.27\text{–}0.30$ (Fig. 5), for which there are typically four or five periods of the electron Shubnikov–de Haas oscillations on curves of $\rho(H)$. For the filaments with $d = 0.21 \pm 0.03\ \mu m$ (Fig. 6), the curves of $\rho(H)$ have only two maxima, which stem from the fact that the a electrons reach the quantum limit at $H^a_{ql} \approx 13$ kOe, and the electrons with the b ellipsoids do the same at $H^b_{ql} \approx 27$ kOe. As the thickness of the single crystals is reduced to $d = 0.15\ \mu m$ (Fig. 7), only a single maximum of the Shubnikov–de Haas oscillations remains—a maximum which results from the departure of the last Landau level from the electron b ellipsoids. All of the other maxima on the curve in Fig. 7 are related to the quantum size oscillations in the longitudinal magnetoresistance. Since the amplitude of the high-frequen-

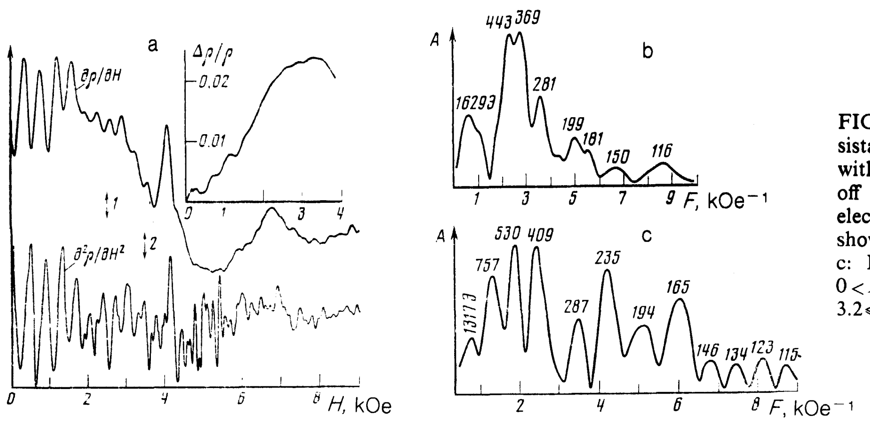

FIG. 4. a: First and second derivatives of the magnetoresistance with respect to the magnetic field for a Bi sample with $d = 0.40\ \mu m$ at 4.2 K. Arrows 1 and 2 show the cutoff fields for Shubnikov–de Haas oscillations from the electrons of the a and b ellipsoids, respectively. The inset shows the initial part of the $\rho(H)$ curve in larger scale. b, c: Fourier spectra of $\partial^2\rho(H)/\partial H^2$ curve in fields $0 < H \leqslant 3.6$ kOe and the $\partial^2\rho(H)/\partial H^2$ curve in fields $3.2 \leqslant H \leqslant 7.6$ kOe, respectively.

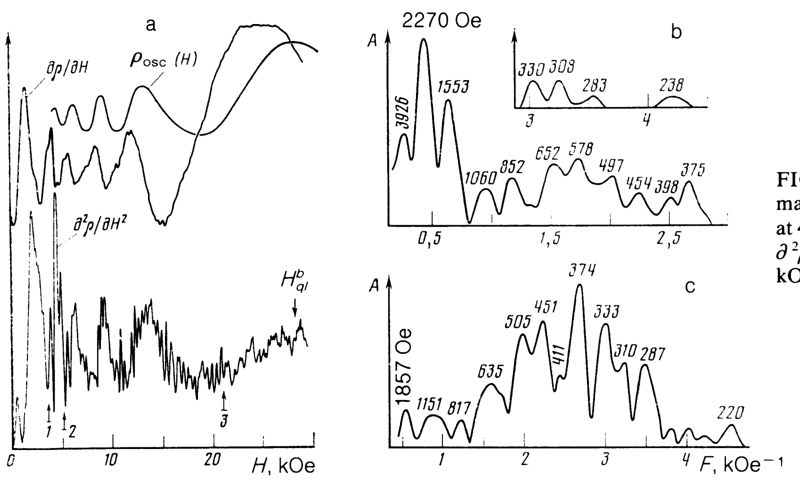

FIG. 5. a: Shubnikov–de Haas and size-effect oscillations in the magnetoresistance of a filamentary Bi sample with $d = 0.27\ \mu$m at 4.2 K. 1—$H^a_{\rm cut}$; 2—$H^b_{\rm cut}$; 3—$H^h_{\rm cut}$. b,c: Fourier spectra of the $\partial^2\rho(H)/\partial^2H$ curve for the respective field intervals $0 < H \leqslant 15$ kOe and $8.1 \leqslant H \leqslant 20.1$ kOe.

cy oscillations (of the second group) is substantially lower than the amplitude of the quantum size oscillations of the first group, the latter are predominant in fields $0 < H \leqslant H^b_{\rm cut}$. At $H > H^b_{\rm cut}$ the amplitude of the quantum size oscillations of the first group falls essentially to zero, and the high-frequency components alone dominate the size–oscillation part of the longitudinal magnetoresistance of filamentary bismuth single crystals. This property of the oscillations with periods of $(0.3–0.9)\Phi_0$, i.e., damping of the amplitude with increasing H which is significantly less pronounced than for the quantum size oscillations of the first group, makes it possible to observe these oscillations in their pure form in stronger magnetic fields.

The high-frequency quantum size oscillations of the longitudinal magnetoresistance, with periods $\Delta\Phi \sim (0.3–0.9)\Phi_0$, thus have the following distinctive features: They begin in weak fields at the same time as the quantum size oscillations of the first group, with $\Delta\Phi \sim (1–4.2)\Phi_0$ (Figs. 5 and 7). Their period is constant in a parallel magnetic field and inversely proportional to the cross-sectional area of the filamentary crystals. The range of magnetic fields in which they exist is far greater than the value of $H_{\rm cut}$ for the electron Shubnikov–de Haas oscillations (Fig. 1). For the thinnest filaments ($d < 0.2\ \mu$m), the quantum size oscillations are also seen in magnetic fields beyond the quantum limit for electrons, but the region in which they exist is limited by the cutoff fields for the hole Shubnikov–de Haas oscillations.

The complexity of the oscillation curves in Figs. 3–7 is a consequence of the complex frequency composition of the quantum size oscillations, as is confirmed by their Fourier spectrum, shown in the same figures. Plotted along the ordinate in the Fourier spectrograms is the relative oscillation amplitude, which determines the contribution of the corresponding oscillation component in the region of magnetic fields under consideration.

It should be recalled that the amplitudes of the peaks on the Fourier spectrograms found for the size oscillations in the curves of $\partial^2\rho(H)/\partial H^2$ by no means characterize the amplitudes of the various oscillations in $\rho(H)$. Differentiating twice sharply increases the relative amplitude of the high-frequency oscillations and also the level of the higher harmonics (primarily the second) in the spectrum of oscillations of each type. Furthermore, the double differentiation causes a substantial weakening of the monotonic trend, with the result that the amplitudes of the false peaks which arise on the Fourier spectrograms because of the finite size of the oscillation interval which is being analyzed decrease or disappear completely. Since "ordinary" quantum size oscillations with $\Delta\Phi \sim (1–4.2)\Phi_0$ dominate in fields $0 < H \leqslant H^b_{\rm cut}$, while high-frequency oscillations with $\Delta\Phi \sim (0.3–0.9)\Phi_0$ dominate in fields $H > H^b_{\rm cut}$, a Fourier analysis of the oscillations curves was carried out separately for each of these two regions of magnetic fields. Table I shows values found for the periods of the oscillation components, ΔH, by numerical calculations. Table II shows the periods in units of the quantum of magnetic flux, Φ_0. It follows from these Fourier spectra (Figs. 4–6) that the error in the determination of the periods of the quantum size oscillations for a given sample through

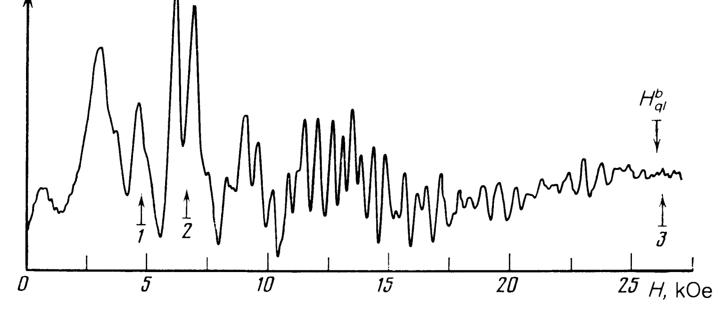

FIG. 6. Quantum-size oscillations in the magnetoresistance of a filamentary Bi sample with $d = 0.21\ \mu$m at 4.2 K. 1—$H^a_{\rm cut}$; 2—$H^b_{\rm cut}$; 3—$H^h_{\rm cut}$.

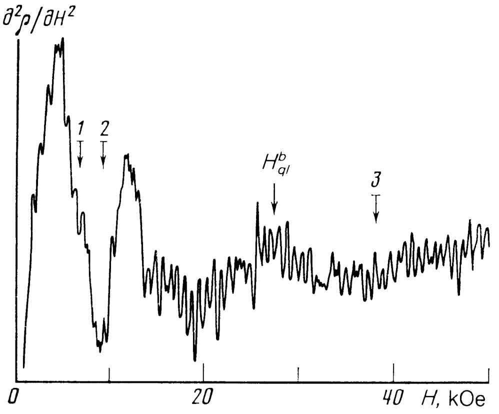

FIG. 7. Quantum-size oscillations in a filamentary Bi sample with $d = 0.15\,\mu$m at 4.2 K. 1—$H^a_{\rm cut}$; 2—$H^b_{\rm cut}$; 3—$H^h_{\rm cut}$. The field interval 14–50 kOe is dominated by high-frequency quantum size oscillations with a period $\Delta\Phi = 0.33\Phi_0$ in the flux ($\Delta H = 780$ Oe).

the use of the curves of $\partial\rho(H)/\partial H$, $\partial^2\rho(H)/\partial H^2$ and through variation of the magnetic-field intervals used in the analysis is less than 5%. The scatter in the values of $\Delta\Phi/\Phi_0$ for a given period for different filaments cannot be linked with an error in the determination of the frequency composition of the quantum size oscillations. It appears to be a consequence of the uncertainty in the determination of the diameter of the samples and possible slight variations in their crystallographic orientation.

A third group is made up of oscillations which are equidistant in a forward magnetic field (Fig. 8). These oscillations differ substantially from those shown in Figs. 3–7 in terms of the magnetic-field interval in which they exist and in terms of their periods, which are far greater than the value $\sim 4\Phi_0$ which is characteristic of the lowest-frequency component of the quantum size oscillations in fields $H < H_{\max}$ (Tables I and II). For a filamentary sample with $d = 1.0$ μm, the curve of $\partial\rho(H)/\partial H$ has equally spaced oscillations with a period of $26\Phi_0$ ($\Delta H = 1370$ Oe; Fig. 8) superimposed on Shubnikov–de Haas oscillations. The amplitude of the oscillations remains essentially constant over the entire range of magnetic fields used (0–50 kOe). Oscillations of the magnetoresistance with a predominant period $\Delta\Phi = 8\Phi_0$ ($\Delta H = 1140$ Oe) in the flux are detected in fields 0–16 kOe for a filament with $d = 0.61$ μm. Because of their large amplitude, they are observed both against the background of the monotonic change in the magnetoresistance and on the curve of $\partial\rho/\rho H$ versus H. Fifteen oscillation periods were detected, with an essentially linear dependence of the index of the extremum on the magnetic field. A Fourier analysis of the $\partial\rho(H)/\partial H$ curve for a filament with $d = 0.61$ μm showed that in addition to the oscillations with a period of $8\Phi_0$ the spectrum of quantum size oscillations of the third group also contains four oscillation components with periods of $(4–18)\Phi_0$ (Table III) and with smaller amplitudes. In Fig. 9, which shows a record of the quantum size oscillations and their Fourier spectrum for a sample with $d = 0.63$ μm, we see that when several frequencies of the third group are predominant the oscillation curves of the longitudinal magnetoresistance have a shape which is far more complex than that of the curves in Fig. 8. The quantum size oscillations of this type are unusual in that they are observed in fields $H \gg H_{\rm cut}$ for electrons and holes superimposed on Shubnikov–de Haas oscillations, where the quantum size oscillations of the first and second groups are absent.

Table III shows the frequency composition of the quantum size oscillations of the third group for several filamentary samples. Shown in parentheses is the index of the oscil-

TABLE I. Periods of the quantum size oscillations (of the first and second groups) in the longitudinal magnetoresistance, ΔH (in oersteds), for filamentary bismuth single crystals at 4.2 K.

d, μm	Period, ΔH, Oe							
0.68	474	322	251	196	150	127	–	–
0.60	–	425	–	264	210	166	–	142
0.58	660	409	296	–	205	–	157	–
0.52	810	–	448	–	291	232	–	191
0.47	814	–	467	–	–	269	–	219
0.43	1077		633	–	–	342	–	276
0.40	1370	860	–	590	443	369	–	–
0.27	2270	–	1553	–	1060	852	–	652
0.24 (a)	3150	–	1800	–	1230	–	–	–
0.24 (б)	3883		1920	1667	1320	1182	–	860
0.23	–	2514	–	–	–	–	1062	–
0.21 (a)	–	3380	–	2020	–	1430	–	–
0.21 (б)	–	3040	2280	–	–	1465	–	–

d, μm	Period, ΔH, Oe							
0.68	100	80	70	–	–	–	–	–
0.60	118	–	–	–	–	–	–	–
0.58	131	114	105	–	–	–	–	–
0.52	164	–	–	–	–	–	–	–
0.47	–	–	157	–	–	114	–	–
0.43	234	–	199	173	–	136	–	–
0.40	281	235	–	194	165	–	123	–
0.27	578	–	497	454	375	330	287	238
0.24 (a)	745	636	–	–	536	455	366	–
0.24 (б)	–	696	590	–	471	373	–	232
0.23	826	–	648	–	555	440	–	345
0.21 (a)	1070	–	738	–	–	570	448	376
0.21 (б)	1063	–	771	711	606	532	443	360

TABLE II. Periods ($\Delta\Phi/\Phi_0$) in the quantum size oscillations (of the first and second groups) in the longitudinal magnetoresistance of cylindrical bismuth single crystals at 4.2 K in arbitrary units.

d, μm	Period, $\Delta\Phi/\Phi_0$							
0.68	4.1	2.8	2.2	1.70	1.30	1.10	–	–
0.60	–	2.9	–	1.80	1.43	1.13	–	0.97
0.58	4.2	2.6	1.88	–	1.30	–	1.00	–
0.52	4.1	–	2.3	–	1.49	1.20	–	0.98
0.47	3.4	–	1.95	–	–	1.12	–	0.91
0.43	3.7	–	2.2	–	–	1.19	–	0.96
0.40	4.1	2.6	–	1.80	1.34	1.11	–	–
0.27	3.1	–	2,1	–	1.46	1.17	–	0.90
0.24(a)	3.4	–	1,96	–	1.34	–	–	–
0.24(b)	4.2	–	2.1	1.81	1.43	1.28	–	0.93
0.23	–	2.5	–	–	1.40	–	1.06	–
0.21(a)	–	2.8	–	1.68	–	1.19	–	–
0.21(b)	–	2.5	1.90	–	–	1.22	–	–

d, μm	Period, $\Delta\Phi/\Phi_0$							
0.68	0.87	0.70	0.61	–	–	–	–	–
0.60	0.80	–	–	–	–	–	–	–
0.58	0.83	0.73	0.67	–	–	–	–	–
0.52	0.84	–	–	–	–	–	–	–
0.47	–	–	0.65	–	–	0.47	–	–
0.43	0.81	–	0.69	0.60	–	0.47	–	–
0.40	0.85	0.71	–	0.59	0.50	–	0.37	–
0.27	0.80	–	0.68	0.62	0.51	0.45	0.39	0.33
0.24(a)	0.81	0.72	–	–	0.58	0.49	0.40	–
0.24(b)	–	0.76	0.64	–	0.51	0.41	–	0.35
0.23	0.83	–	0.65	–	0.55	0.44	–	0.34
0.21(a)	0.89	–	0.61	–	–	0.47	0.37	0.31
0.21(b)	0.88	–	0.64	0.59	0.50	0.44	0.37	0.30

lation component, in order of decreasing amplitude of the component. Although the oscillation part of the magnetoresistance of these filaments contains up to 11 frequencies (of the third group), the values of their relative periods $\Delta\Phi/\Phi_0$ agree well with the corresponding periods found for samples of other diameters. Analysis of the thickness dependence of the low-frequency oscillations in the magnetoresistance with $\Delta\Phi \gg \Phi_0$ shows that the periods of the constituent oscillation components of these oscillations are inversely proportional to the cross-sectional area of the filamentary samples. This property is convincing proof that such oscillations in the magnetoresistance fall in the class of size oscillations. It should be noted that low-frequency quantum size oscillations can be seen best in filaments with a thickness of 0.4–1.0

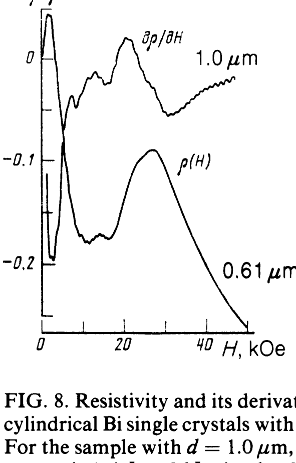

FIG. 8. Resistivity and its derivative with respect to the magnetic field in cylindrical Bi single crystals with thicknesses of 0.61 and 1.0 μm at 4.2 K. For the sample with $d = 1.0$ μm, quantum size oscillations with an average period $\Delta\Phi = 26\Phi_0$ in the flux $[(\Delta H)_{cp} = 1370$ Oe$]$ can be seen against the background of the Shubnikov–de Haas oscillations.

μm, which have the maximum resistance ratio $\rho_{300}/\rho_{4.2}$. Table III shows, for the example of a filamentary sample with $d = 0.24$ μm, that the thinnest bismuth filaments exhibit low-frequency quantum size oscillations with a maximum period $\sim 10\Phi_0$ in fields up to 50 kOe.

The low-frequency quantum size oscillations are thus manifested in the weakest magnetic fields, along with the quantum size oscillations of the first and second groups, but they continue to be seen in fields above H^e_{cut} and H^h_{cut} and above the quantum limit for b electrons. There is no unambiguous correlation between the diameter of the filamentary sample and the magnetic-field region over which the low-frequency quantum size oscillations exist. In a first approximation, the low-frequency quantum size oscillations are equidistant in the magnetic field, and their periods depend on the diameter in accordance with $\Delta H \sim d^{-2}$, lying in the range $(2–26)\Phi_0$. The smallest period of the low-frequency quantum size oscillations is most often $(3.5–4.2)\Phi_0$; the amplitude of the low-frequency quantum size oscillations depends primarily on the quality of the interior and surface of the filamentary single crystals. In samples with $d = (0.6–1.0)$ the maximum amplitude is that of oscillation components with periods of $(8–26)\Phi_0$; components with $\Delta\Phi < 10\Phi_0$ become predominant as the diameter of the filament is reduced.

In this study we did not carry out a detailed analysis of the temperature dependence of the oscillation amplitude.

5. DINGLE OSCILLATIONS AND "FLUX-QUANTIZATION" OSCILLATIONS FROM THE ELECTRON FERMI SURFACES OF BISMUTH

It was shown above that in magnetic fields $0 < H \lesssim H^b_{cut}$ quantum size oscillations with periods in the flux lying in the

TABLE III. Periods of the quantum size oscillations (of the third group) in the longitudinal magnetoresistance of filamentary bismuth samples at 4.2 K.

$d=0.63\ \mu m$				$d=0.62\ \mu m$				$d=0.61\ \mu m$		$d=0.24\ \mu m$	
$H<H_{max}$		$H>H_{max}$		$H<H_{max}$		$H>H_{max}$		$H>H_{max}$		$0<H\leqslant 50$ kOe	
ΔH, Oe	$\Delta\Phi/\Phi_0$	ΔH, Oe	$\Delta\Phi/\Phi_0$	ΔH, Oe	$\Delta\Phi/\Phi_0$	ΔH, Oe	$\Delta\Phi/\Phi_0$	ΔH, Oe	$\Delta\Phi/\Phi_0$	ΔH, Oe	$\Delta\Phi/\Phi_0$
–	–	3430	25.8(2)	–	–	3350	24.3(6)	–	–	–	–
2200	16.5(1)	2340	17.6(4)	–	–	2650	19.2(4)	2550	18.0(2)	–	–
–	–	–	–	–	–	2030	14.7(1)	–	–	–	–
–	–	1580	11.9(1)	–	–	1600	11.6(5)	1660	11.7(3)	–	–
–	–	–	–	–	–	1330	9.6(2)	–	–	9440	10.3(1)
–	–	1080	8.1(3)	1120	8.1(2)	1100	8.0(3)	1144	8.1(1)	–	–
–	–	–	–	–	–	920	6.7(7)	–	–	5800	6.3(5)
763	5.7(5)	850	6.4(5)	–	–	820	5.9(8)	826	5.8(4)	–	–
–	–	750	5.6(6)	–	–	–	–	–	–	–	–
–	–	640	4.8(10)	–	–	655	4.7(10)	–	–	–	–
–	–	582	4.4(8)	–	–	600	4.3(9)	–	–	–	–
544	4.1(2)	–	–	–	–	–	–	570	4.0(5)	3880	4.2(4)
–	–	495	3.7(7)	440	3.2(6)	–	–	–	–	3140	3.4(3)
343	2.6(6) *	380	2.9(9)	–	–	–	–	–	–	2550 **	2.8(2) *
–	–	270	2.0(11)	–	–	–	–	–	–	–	–
228	1.71(4) *	–	–	250	1.82(4) *	–	–	–	–	1660 **	1.8(6) *
200	1.50(3) *	–	–	194	1.40(5) *	–	–	–	–	1290 **	1.4(7) *
166	1.25(7) *	–	–	156	1.13(1) *	–	–	–	–	–	–
143	1.07(8) *	–	–	123	0.90(3) *	–	–	–	–	920 **	1.0(8) *

*Oscillations of the first group, observed in filamentary bismuth samples at 4.2 K.
**At $0<H\leqslant 14$ kOe.

interval $\Delta\Phi\sim(1\text{–}4.2)\Phi_0$ dominate. It follows from Table I–III that in the interval $(1\text{–}4.2)\Phi$ there are six characteristic periods, with values, in units of $\Delta\Phi/\Phi_0$, of 3.7 ± 0.5, 2.7 ± 0.2, 2.0 ± 0.2; 1.7 ± 0.1, 1.4 ± 0.1, and 1.1 ± 0.1.

The oscillations with a period $(1.1\pm0.1)\Phi_0$ which were first observed in Refs. 4, 5, 20, and 21, are predominant on the $\rho_{\parallel}(H)$ curve in the weakest fields and fade rapidly in the field interval $0<H\lesssim H_{cut}$. These oscillations should be classified as the "flux-quantization" oscillations which were predicted theoretically in Refs. 9–11. The other periods, except the first, agree well with the periods of Dingle oscillations calculated for electrons for cylindrical bismuth single crystals in Refs. 13 and 14. The theoretical calculations of the periods of the quantum size oscillations in those studies were carried out exclusively for the electrons on the a ellipsoid (see the inset in Fig. 1), whose extension direction makes a small angle ($\sim20°$) with the axis of the filamentary sample and whose intersection with the plane perpendicular to the axis of the filament and for which the area of the intersection with the plane perpendicular to the axis of the filament is nearly the minimum area. It has been suggested that for the electrons of this group the conditions for size quantization in cylindrical single crystals hold better than for the electrons of the two other equivalent b ellipsoids, with a cross-sectional area $S_b\approx2S_a$, for which the values of the extremal Fermi momenta in the plane perpendicular to the axis of the filament are larger. However, since there are twice as many electrons in the b ellipsoids as in the a ellipsoid, and since the extremal Fermi momenta differ by a factor of only 1.4, it can be assumed that the b electrons can make an appreciable contribution to the oscillations in the magnetoresistance. The large number of oscillatory components in the quantum size oscillations in the magnetoresistance in fields $0<H<H_{max}$ may be a consequence of the contribution of the electrons of b ellipsoids to the oscillations. Evidence in favor of this suggestion comes from the fact that the quantum size oscillations with periods $(1.4\text{–}4.2)\Phi_0$ for most of the samples with $d<0.7\ \mu m$ in fields equal to H^a_{cut} still have a substantial amplitude (Figs. 3–7) and are detected up to fields as strong as the cutoff field for the electrons of the b ellipsoids, which are $H^b_{cut}\approx1.4H^a_{cut}$. In contrast, the theoretical calculations of Refs. 7–11, 13, and 14 state that the amplitude of the quantum size oscillations from the corresponding group of carriers should vanish in a magnetic field equal to the cutoff field. Using the expression derived in Ref. 14 to determine the periods of the quantum size oscillations of cylindrical bismuth single crystals, we can calculate the periods of the Dingle quantum size oscillations from electrons of the b ellipsoids. In units of $\Delta\Phi/\Phi_0$, these periods are 1.71, 1.47, 1.24, 1.18, and 1.15. We see that the frequency spectra of the quantum size oscillations from the electrons of the a and b ellipsoids overlap significantly; the effect is to

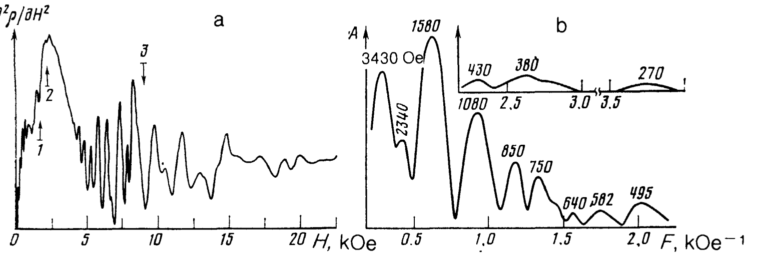

FIG. 9. a: Quantum-size oscillations in the longitudinal magnetoresistance of a Bi filament with $d=0.63\ \mu m$ at 4.2 K. 1—H^a_{cut}; 2—H_{cut}; 3—H^b_{cut}. b: Fourier spectrum of the quantum size oscillations in fields $3.7\leqslant H\leqslant22$ kOe for a sample with $d=0.63\ \mu m$.

complicate an unambiguous separation of the contribution of each of the electron groups to the oscillatory part of the magnetoresistance.

6. DINGLE OSCILLATIONS FROM THE HOLE PART OF THE FERMI SURFACE OF BISMUTH

The high-frequency quantum size oscillations (of the second group; Figs. 4–7) should be put in this category. These oscillations contain four to seven (most often, five) frequencies, with periods which are distributed almost uniformly over the interval $(0.3–0.9)\Phi_0$ (Tables 1 and 2). Are some of these frequencies associated with the double differentiation of the $\rho(H)$ curve which we carried out in order to distinguish the size-dependent oscillatory part of the longitudinal magnetoresistance? To answer this question, we carried out a Fourier analysis of the curves of both $\partial\rho(H)/\partial H$ and $\partial^2\rho(H)/\partial H^2$ for most of the filamentary bismuth samples. We found that the frequency spectra of these two curves are qualitatively the same (Fig. 4). The amplitude of the high-frequency quantum size oscillations depends on H in a complicated way. In fields $0<H<H^b_{\rm cut}$ the amplitude of the oscillations with $\Delta\Phi\sim(0.3-0.9)\Phi_0$ decreases with increasing oscillation frequency, but for $H>H^b_{\rm cut}$ this behavior changes. For most of the filamentary bismuth samples the region in magnetic field in which the high-frequency quantum size oscillations exist overlaps the region of electron Shubnikov–de Haas oscillations and lies in the interval $0<H\lesssim H^h_{\rm cut}$ ($H_{\rm cut}d = 5.6\ \mu$m·kOe; Fig. 1). This circumstance is responsible for the complicated way in which the amplitude of these oscillations falls off with increasing H in comparison with the electron Dingle oscillations. Shubnikov–de Haas oscillations from T holes appear in essentially all of the samples in fields at which the amplitude of the high-frequency quantum size oscillations vanishes.

The higher frequency of the quantum size oscillations of the second group is a consequence of the small distance between the size-effect levels, $\Delta\varepsilon^h_{n+1,n}$, itself a consequence of the large effective mass which is characteristic of the T holes. The small values of the distance between the hole size-effect levels also explains the relatively low amplitude of the high-frequency quantum size oscillations.

In the crystallographic orientation characteristic of cylindrical bismuth single crystals the conditions for size quantization for T holes are the least favorable, because the extremal Fermi momentum in the plane perpendicular to the axis of the sample is close to the maximum value. Nevertheless, estimates show that in filaments $\sim 0.4\,\mu$m thick the distance between the size-effect energy levels closest to the Fermi surface for the T holes is $\Delta\varepsilon^h_{n+1,n}\approx 1$ meV, i.e., essentially the same as $\Delta\varepsilon^e_{n+1,n}$ for the discrete electron levels in samples with $d\approx 1.0\,\mu$m, where quantum size oscillations from electrons can be reliably detected. In filamentary single crystals $0.2–0.3\,\mu$m thick the energy gap between the hole size-effect levels is ~ 2 meV. Clearly, samples of very high quality will be required in order to see the quantum size oscillations associated with such a small spacing in the energy spectrum.

According to the data of Ref. 22, the probability for the specular reflection of holes from a surface is substantially lower than that for electrons. Consequently, high-frequency quantum size oscillations are not detected for all of the filamentary samples but only for those which have the best bulk and surface characteristics.

7. QUANTUM OSCILLATIONS ASSOCIATED WITH MAGNETIC SURFACE LEVELS

It was shown above that the Dingle oscillations from "bulk" electrons and holes are detected in most of the filamentary bismuth samples up to fields equal to $H_{\rm cut}$ for the corresponding carrier group. For filaments with $d = 0.6–1.0\ \mu$m, low-frequency quantum size oscillations are detected at fields up to 30–50 kOe (Figs. 8 and 9—i.e., up to fields three to nine times greater than $H^h_{\rm cut}$ for holes and tens of times greater than the cutoff fields $H^a_{\rm cut}$ and $H^b_{\rm cut}$ for electrons. Oscillations of this type in the magnetoresistance cannot be identified as the quantum size oscillations observed in Refs. 2–5, since the latter were observed only in fields preceding the electron Shubnikov–de Haas oscillations. There is a difficulty in determining the physical nature of the size-effect oscillations with periods $\Delta\Phi\gg\Phi_0$ which are characteristic of some of the bismuth samples: essentially all of the existing theoretical studies predict quantum size oscillations in the magnetoresistance only at fields at which the carrier Larmor radius r_H exceeds the thickness of the sample. Calculations by Mankin[13] and Nedorezov[14] show that the maximum period of the Dingle oscillations in the magnetoresistance for the bismuth filaments in the orientation which we studied would be $\sim 2.6\Phi_0$.

The low-frequency quantum size oscillations with periods $\Delta\Phi\sim(4–26)\Phi_0$ observed in the present experiments (Table III) thus disagree both in terms of the magnetic-field interval in which they exist and in terms of their periods with the results of other experimental and theoretical studies of quantum oscillation phenomena in cylindrical conductors.

To determine the nature of the low-frequency quantum size oscillations, let us make use of their primary distinguishing feature: They exhibit an "independence" both with respect to the Dingle oscillations and with respect to the Shubnikov–de Haas oscillations associated with the bulk charge carriers. This circumstance indicates that the quantum size oscillations of this type are not related to bulk carriers and probably stem from the existence of magnetic surface levels.[12] The existence of size oscillations in the magnetoresistance is convincing evidence for the specular nature of the reflection of the charge carriers from the surface of the filamentary bismuth single crystals studied in these experiments. This conclusion in turn means that magnetic surface levels may exist in these crystals.

As we mentioned in the Introduction, a similar oscillatory dependence of the longitudinal magnetoresistance of cylindrical conductors was studied in Refs. 9–11, where it was shown that the spectrum of magnetic surface levels in quantum size cylindrical samples is quite different from that in a bulk sample. According to Refs. 9–11, the motion of grazing electrons in a narrow layer near the surface in a weak field, $r_H\gg R$, leads to oscillations in thermodynamic and kinetic properties with a universal period $\Delta\Phi=\Phi_0$. This period should be independent of the particular carrier dispersion law. The experimental observation of multifrequency quantum size oscillations with periods $\Delta\Phi\gg\Phi_0$ and their existence in fields $H\gg H_{\rm max}$ indicate that the physical nature of the low-frequency quantum size oscillations is more complicated than that of the oscillations studied in Refs. 9–11.

Analysis of the periods in the magnetoresistance oscillations of electrons in bismuth filaments in fields $H<H_{\rm max}$ has revealed six characteristic frequencies over the interval

$(1–4.2)\Phi_0$ and has also revealed that the period $\Delta\Phi = (3.7 \pm 0.5)\Phi_0$ does not agree with the results of the calculations in Refs. 13 and 14. The experimental results shown in Table III explain this discrepancy and indicate that quantum size oscillations with a period $\sim 4\Phi_0$ differ in physical nature from the quantum size oscillations studied in Refs. 13 and 14 and apparently stem from magnetic surface levels.

In bismuth filaments with $d \sim 0.4–1.0\ \mu m$, the amplitude of the low-frequency quantum size oscillations in fields $H > H_{max}$ is some four to six times higher than that of the Dingle quantum size oscillations at $H < H_{max}$. In general, as H is increased the low-frequency quantum size oscillations exhibit a clearly defined beat nature (Fig. 9), and their amplitude decreases; i.e., their behavior in a magnetic field is similar to that of quantum size oscillations from bulk carriers.

We do not rule out the possibility that the physical nature of the low-frequency quantum size oscillations might have some different explanation, e.g., one based on a fluctuation model.[23,24]

8. CONCLUSION

The theory worked out on the quantum size oscillations in Refs. 7, 9–11, 13, and 14 was restricted to weak fields, in which the role of the magnetic field reduces to one of simply shifting the system of quantum size-effect energy levels which exist in a filamentary sample. For this reason, the use of the results of these calculations to analyze experimental data obtained in stronger fields is not strictly correct. In strong fields, the situation may be complicated by the disruption of the equidistant arrangement of the oscillation peaks in a forward magnetic field (the period acquires a field dependence) and by the oscillations of the Fermi level, with the result that frequency modulation of the quantum size oscillations should set in. In principle, each of these effects can complicate Fourier analysis of the experimental oscillatory dependence of the magnetoresistance, but at present it does not seem possible to avoid these difficulties.

A study of the longitudinal magnetoresistance of bismuth single-crystal filaments has shown that in samples with $d \leqslant 1\ \mu m$ there are two types of quantum size oscillations in the magnetoresistance, which differ in physical nature. Classified as oscillations of the first type are those which stem from the discrete energy spectrum of the bulk charge carriers.[7,9–11,13,14] The reason for the complex oscillations of this type in the magnetoresistance is the fact that bismuth has several nonequivalent constant-energy surfaces, each of which makes its own contribution to the quantum size oscillations in the magnetoresistance. The results of the present experiments show that both the electrons at the L point in the Brillouin zone and holes at the T point participate in the formation of the quantum size oscillations of the first type in bismuth filaments.

One possible reason for the appearance of quantum size oscillations of the second type in the magnetoresistance might be the appearance of magnetic surface levels in filamentary samples. These surface levels would be associated with charge carriers which are moving along "jumping" trajectories over the surface of the filament. Magnetic surface levels characterizing stationary states of electrons which are moving along "jumping" trajectories play the same role for the carriers of this group as Landau levels play for charge carriers in bulk crystals. As in the case of size oscillations from bulk carriers, the apparent reason for the complex frequency spectrum of quantum size oscillations in the magnetoresistance due to the surface levels is the contribution of carriers which belong to the electron and hole parts of the Fermi surface of bismuth.

We need to stress that the quantum size oscillations with periods $(1–2.6)\Phi_0$ and $(0.3–0.9)\Phi_0$ are not localized in the field intervals from 0 to H^e_{cut} and H^h_{cut}, respectively, as has been suggested in some theoretical papers.[7–11,13,14] A spectral analysis reveals quantum size oscillations of electrons and holes even in fields above the corresponding cutoff field, as is illustrated by the results in Figs. 5 and 7. As a rule, the amplitude of the quantum size oscillations at $H > H_{cut}$ is lower than the amplitude of the oscillations in fields below H_{cut}.

Studies of the quantum size oscillations in the resistance in a magnetic field are considerably more convenient and informative than studies of the quantum size oscillations which stem from the thickness of the sample at $H = 0$, since in the former case it becomes possible to separate the contributions made to the quantum size oscillations in the magnetoresistance by different groups of charge carriers, both by varying the magnetic field and by analyzing the data (by Fourier analysis, etc.) for the "size-effect" oscillatory part of the $x(H)$ curve. Furthermore, the specific manifestations of quantum size oscillations in magnetic fields give rise to new physical phenomena which are of independent, possibly practical interest.

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