

Review Article

phys. stat. sol. (a) **1**, 199 (1970)

Subject classification: 1.4; 13.1

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Quantum Size Effect—Present State and Perspectives of Experimental Investigations

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1. Introduction

The quantum size effect (QSE) experimentally observed in 1965 [1, 2] consists in the following: In films whose thickness d is comparable with the effective De-Broglie wavelength of the carriers, λ , the effect of the size quantization of the energy of the transverse motion of electrons may occur. The energy spectrum of such a system is described (in addition to the band number) by means of two continuous and one discrete quantum numbers, and presents a system of subbands, each of which corresponds to a fixed value of the discrete quantum number. Under these circumstances, the minimum energy of the carriers is different from zero and a function of the film thickness.

The presence of a discrete quantum number leads to a non-monotonous dependence of the density of electron states on energy, $g(E)$. In the particular case of quadratic dispersion this function shows flat steps.

The peculiar properties of the energy spectrum of thin films result in an anomalous behaviour of several fundamental macroscopic characteristics of solids (thermodynamic, kinetic, etc.), which enables one to speak about the so-called "film" state of the substance.

The purely theoretical investigations, preceding the experimental observation of the quantum size effect, are referred to in [3] to [9].¹⁾ In [4], in particular, it is pointed out that the quantum size effect should not be expected in markedly degenerate systems. In this work, as well as in [5], some conditions essential for the existence of the quantum size effect are formulated.

It should here be pointed out that the actual presence of the quantum size effect, which in principle follows from the quantum theory of solids, was not obvious on account of fairly rigid conditions for its realization formulated in [4, 5].

In addition to the main condition $d \sim \lambda$, other conditions permitting the existence of the quantum size effect may be summed up in a requirement for a fairly small washing out of the levels, $\delta E = \hbar/\tau \ll E_0$ (where τ is the relaxation time, taking into account scattering both in volume and on the surface; $E_0 = \pi^2 \hbar^2/2 m^* d^2$ is the zero energy), a fairly high uniformity of the film as to its thickness, and, as required for the observation of oscillation effects in degenerate systems, in the obvious requirement $E_0 > kT$.

The condition $d \sim \lambda$ selects a material with small effective mass and low Fermi energy E_F . Inequality $\hbar/\tau \ll E_0$ actually means a requirement for the free path length l ($l \gg d$), where l is a free path in a bulk sample of the same quality as that of the film.

The above-mentioned conditions are fulfilled in bismuth where $\lambda \approx 10^{-5}$ cm, ($E_F \approx 10^{-2}$ eV, $m_z^* \approx 10^{-2} m_0$, the z -axis being in direction of the trigonal crystal axis), $l \approx 1$ mm at 4.2 °K.

2. Samples under Investigation

The main results of the investigations of the quantum size effect have been obtained on thin layers of bismuth, condensed in vacuum on a mica (muscovite) substrate [1, 2, 10, 11, 17, 20, 25, 33, 34].

The purity of the starting bismuth was 99.9999%. The material was usually deposited from a graphite or tantalum vaporizer on a hot substrate (heated up to a temperature of 70 to 90 °C) in vacuum $\approx 10^{-6}$ Torr.

The condensation rate was usually equal to ≈ 50 Å/min.

The structure of the bismuth films on mica was single-crystalline, the trigonal axis of the crystal being perpendicular to the film plane (Fig. 1a). The average distance between the exit point of the dislocations on the surface (size of the grains) equals 3 to 5 μ m (Fig. 1b).²⁾

The electrical properties of the samples are characterized by the following figures: At 4.2 °K the concentration and mobility of carriers are $n \approx 3 \times 10^{17}$ cm $^{-3}$ and $\mu \approx 20000$ cm 2 /Vs ($d \approx 2000$ Å), respectively. At 300 °K resistivity of the films is higher than that of bulk samples by as much as ≈ 1.5 times.

In order to obtain samples of another crystallographic orientation and to make optical measurements in the infrared region the bismuth films were also condensed on KBr substrates using conditions similar to those described above [22].

The bismuth samples condensed on KBr substrates were essentially single-crystals with their binary axes perpendicular to the film plane (Fig. 1c).

The electrical characteristics of the samples thus obtained are similar to those of the bismuth films deposited on mica.

¹⁾ See also the review article by Tavger and Demikhovskii [9b].

²⁾ For details concerning the epitaxial growth of bismuth films on mica see [10] and [40].

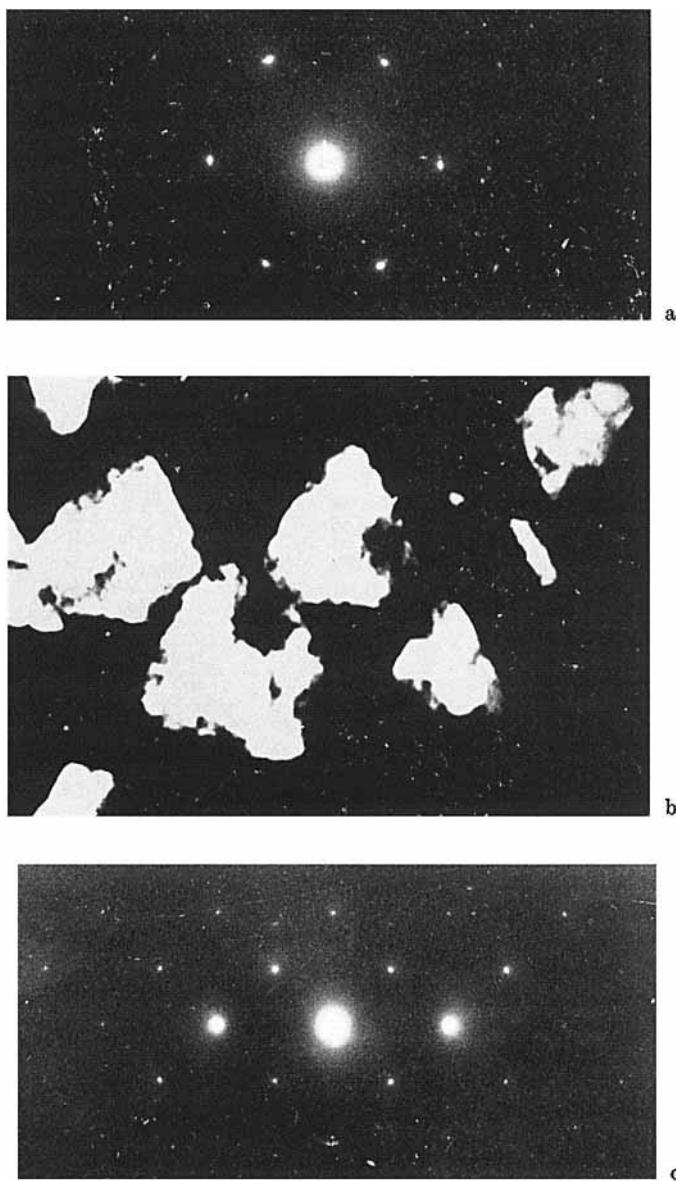


Fig. 1. a) Electron diffraction pattern for a bismuth film ≈ 1300 Å thick condensed on mica.
b) Electron-microscopical pattern of a single-crystalline bismuth film ≈ 1300 Å thick condensed on mica ($18000\times$).
c) Electron diffraction pattern for a bismuth film ≈ 1000 Å thick condensed on KBr

The thickness of the samples was determined from the time of the condensation of bismuth at constant condensation rate. The independence of the deposition rate from the thickness was controlled by a method of multibeam interferometry within a thickness range of approximately 5000 to 70 Å. The absolute value of the condensation rate was determined in a region of large thicknesses with an accuracy of not less than 10%.

3. Kinetic and Galvanomagnetic Phenomena

The presence of a discrete quantum number and peculiarities in the density of states connected therewith should lead to an oscillatory dependence of the thermodynamic and kinetic characteristics of degenerate systems on the film thickness [3, 4].

These oscillations are connected with a variation in the number of subbands located below the Fermi level when the film thickness is varied and the density of states on the Fermi surface changes abruptly provided that the bottom of a sequential subband coincides with the Fermi energy.

The first attempt of detecting the quantum size effect consisted in the measurement of the dependence of the resistivity ϱ , Hall constant R_H , and magnetoresistance $\Delta\varrho/\varrho$ on the thickness of the bismuth films [1, 10].

The samples under test have been obtained by vacuum deposition of bismuth on hot mica. A single technological cycle comprised the simultaneous deposition treatment of 48 samples of different thicknesses (12 samples were placed on each of 4 substrates). The thickness was varied stepwise or gradually ("wedge-type variation").

Measurements of ϱ , $\Delta\varrho/\varrho$, and R_H were made by the conventional compensation method in a magnetic field $H < 1$ kG, perpendicular to the film plane.

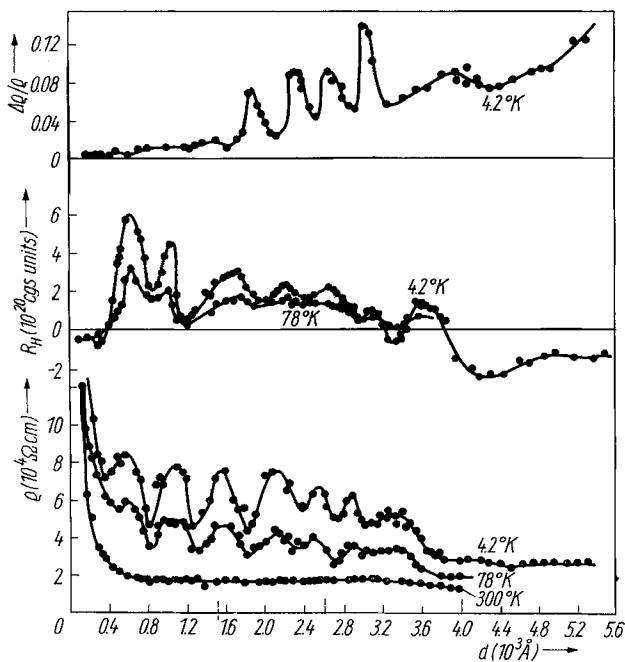


Fig. 2. Dependence of resistivity, Hall coefficient, and magnetoresistance on the thickness of bismuth films condensed on mica (the samples with a stepwise thickness variation)

Fig. 2 represents typical results as obtained by measuring the thickness dependence of ρ , R_H , and $\Delta\rho/\rho$ in a thickness range $d \approx 200$ to 5000 Å at temperatures 4.2, 78, and 300 °K on samples with stepwise thickness variation. The main peculiar feature of the shown curves consists in the presence of oscillations. The oscillation amplitude decreases as the film thickness increases and temperature rises. The oscillations of $\rho(d)$ and $\Delta\rho/\rho(d)$ are found to be in opposite phases. The oscillation period Δd in Fig. 2 equals to ≈ 450 Å and is independent of temperature from 4.2 to 78 °K.

Further investigations have shown that the period of the oscillations can be varied in a range of $\Delta d \approx 250$ to 450 Å by changing the condensation conditions of the films. The main factor determining the value of Δd is probably the substrate temperature. Fig. 3 (the uppermost curve) shows the oscillation dependence $\rho(d)$ having the period $\Delta d \approx 250$ Å [11].

It is also necessary to point out that for the films obtained under conditions corresponding to Fig. 3 ($T_{\text{substrate}} = 85$ °C), a dispersion of the value Δd is observed. The period of $\rho(d)$ increases from ≈ 100 to 150 Å within a region $d \approx 800$ to 1400 Å in a short variation range d (1400 to 1800 Å) up to a value of 250 Å which does not vary until the oscillations are attenuated at great sample thickness. It is not excluded that the dependence $\Delta d(d)$ is caused by the appearance of additional harmonics of electron oscillations in the region $d < 2000$ Å.

Let us interpret the experimental results. The presence of oscillations and the fact that their amplitude decreases with increasing thickness and temperature indicate the existence of the quantum size effect. The fact that the oscillatory dependencies of $\rho(d)$ and $\Delta\rho/\rho(d)$ are found to be in opposite phases, does also confirm the above interpretation. As a matter of fact, a decrease in mobility with an abrupt variation of the density of states on the Fermi surface simultaneously results in an increase in the value of ρ and in a decrease in the value of $\Delta\rho/\rho$.

Furthermore, the oscillation period allows the value of the Fermi momentum $P_z^{(F)}$ in the direction of the trigonal axis to be determined, as well as the value of the component of the effective mass of the carriers in the same direction (m_z^*).³⁾ The obtained values $P_z^{(F)} \approx 7.8 \times 10^{-22}$ gcm/s and $m_z^* \approx 0.025 m_0$ ($\Delta d = 400$ Å; $E_F = 25$ meV) practically agree with similar characteristics of the electron part of the Fermi surface in bulk bismuth [12].⁴⁾

These arguments lead us to the conclusion that in the described experiments the size quantization of the electron part of the energy spectrum has taken place.

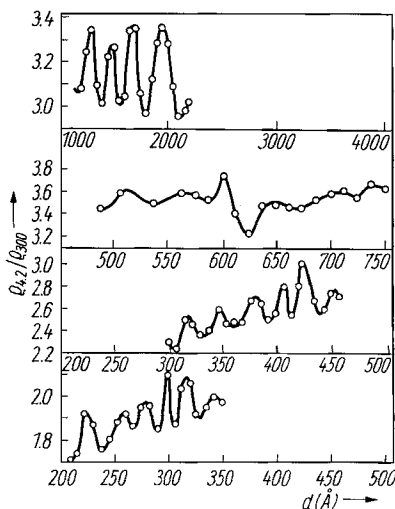


Fig. 3. Dependence of $\rho_{4.2}/\rho_{300}$ on the thickness of bismuth films on mica ("wedge" type)

³⁾ E_F may be determined independently from characteristics of the tunnel effect (see Section 8).

⁴⁾ Let us emphasize that the above-mentioned variation of Δd in a range of 250 to 450 Å does not substantially affect the evaluations thus made.

In a detailed investigation of the dependence $\varrho(d)$ in a region of small thicknesses (about 800 to 200 Å) carried out later [11], it was observed an oscillatory nature of the above-mentioned curve with a very small period gradually varying from $\Delta d \approx 25$ Å for $d \approx 200$ Å to $\Delta d \approx 40$ Å for $d \approx 800$ Å.

Fig. 3 illustrates the variation $\varrho_{4.2}/\varrho_{300}(d)$ for a number of samples of different thicknesses (wedge-type) as obtained under the same operation conditions.

In the region of great thicknesses ($d > 1800$ Å) oscillations are observed with a constant period $\Delta d \approx 250$ Å, connected with the electron part of the Fermi surface.

In the region of small thicknesses (200 to 500 Å) clearly visible high-frequency oscillations with a period $\Delta d \approx 25$ Å appear.

Finally, within a region of thicknesses ≈ 400 to 800 Å low-frequency and high-frequency oscillations are observed simultaneously.

Using the arguments which have been stated earlier for interpreting the electron oscillations (in particular, consistency of the experimental values m_z^* and $P_z^{(F)}$ with the data obtained for bulk bismuth) and also the simultaneous existence of two periods in the intermediate region of thicknesses, it is possible to conclude that the observed high-frequency oscillations are apparently connected with the hole part of the Fermi surface.

It should be pointed out that a difference in the periods of the electron and hole oscillations results from the anisotropic properties of bismuth. This, in particular, explains the fact that the theory of kinetic phenomena in a thin film [13], using an isotropic model of a semi-metal, which correctly reflects the non-monotonous nature $\varrho(d)$, does not allow the separation of the electron and hole type oscillations. The theory [26] using an anisotropic model is in good agreement with experimental data.

The results of the measurements described concerning the electron part of the spectrum were confirmed in the works [14] and [15], where thickness oscillations of the resistivity of bismuth films of the same orientation, corresponding to a large value of Δd ($\Delta d_e \approx 400$ Å [14] and $\Delta d = 360$ Å [15]), were also observed.

A similar oscillatory dependence $\varrho(d)$ was observed by the authors [16] on antimony films. In conformity with a greater value of the electron effective mass in antimony, the value Δd was found to be equal to ≈ 30 Å.

Another peculiarity in the behaviour of the kinetic coefficients in a thin film is referred to in work [41]. When an electric field is applied to a specimen the electron gas is heated up. In this case the current-voltage characteristic of thin films should show peculiar features each time when the energy absorbed by an electron on its free path, becomes equal to the separation of the quasi-discrete levels in the film.

This effect has probably been experimentally observed in [42] on Bi films. In this case, however, it should be noted that the amplitude of the observed peculiarities was comparable with the scattering of the experimental points.

Size oscillations of resistance and Hall mobility of degenerate n-type InSb have been observed in [43]. It was found that a decrease in film thickness removes the degeneracy.

4. Temperature Dependence of Resistivity

The temperature dependence of the resistivity of bismuth films of different thicknesses [17] was determined in order to study further the kinetics of the carriers under conditions of the quantum size effect.

The measurements were performed in a temperature range 4.2 to 200 °K. The temperature was varied by means of a heating appliance placed into a helium-filled Dewar vessel.

The temperature was raised at a rate not exceeding 3 deg/min. The temperature measurements were made with the aid of a platinum thermistor with an accuracy of ≈ 1 °K (at $T > 15$ °K).

The measurements of $\varrho(T)$ were performed with the use of a compensation method in the course of a continuous variation of temperature.

The measurements of the temperature dependence of the resistivity of bismuth films showing oscillations of the kinetic coefficients with thickness have led to the observation of a number of important peculiarities.

First of all, an activation nature of the dependence $\varrho(T)$ is observed. Fig. 4 illustrates the linear dependence $\ln[(\varrho_{4.2} - \varrho_T)/\varrho_{4.2}] = f(1/T)$, where $\varrho_{4.2}$ is the resistivity at 4.2 °K, ϱ_T the resistivity at a current value of temperature.

The presence of an activation energy, following from the linear characteristic of the dependence $\ln[(\varrho_{4.2} - \varrho_T)/\varrho_{4.2}] = f(1/T)$ is not a trivial fact for bismuth. In the temperature range studied (4.2 to 200 °K) the thermal emission of electrons from the extremum of the valence band, separated from the conduction band by a forbidden energy gap of 20 meV, is negligibly small. Yet the transition from the other extremum of the valence band into the conductivity band should not be of an activation nature owing to the overlapping of the bands.

The following important fact is that the angle of inclination of the straight lines $\ln(1 - \varrho_T/\varrho_{4.2}) = f(1/T)$ depends on the film thickness.

Fig. 5 shows the dependence of tangent of the inclination angle of the above-mentioned straight lines ($\operatorname{tg} \alpha$) on the film thickness. It is obvious that the dependence $\operatorname{tg} \alpha = f(d)$ shows an oscillatory nature. The amplitude of the oscillations decreases as the thickness increases. The distance between the extrema somewhat increases as d increases, and coincides in the region of small thicknesses with the period of oscillations of the kinetic coefficients.

These facts may be interpreted as follows: As it has been already pointed out, the variation in the resistivity in the temperature range studied is caused by the thermal emission of electrons from the extremum of the valence band into the conductivity band. Under the conditions of the quantum size effect, the density of states on the bottom of a free subband in the vicinity of the Fermi level undergoes an abrupt change, which provides in the system some analogue

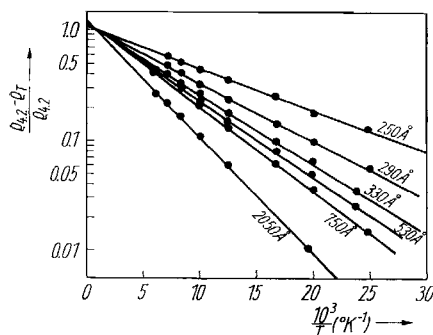


Fig. 4. Dependence $\ln(\varrho_{4.2} - \varrho_T)/\varrho_{4.2} = f(1/T)$ for bismuth films condensed on mica

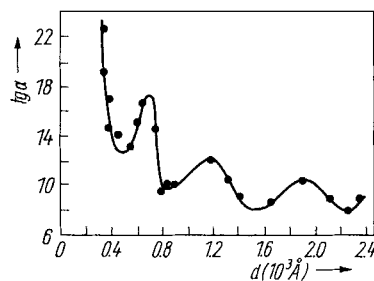


Fig. 5. Dependence of the tangent of the inclination angle for straight lines $\ln(\varrho_{4.2} - \varrho_T)/\varrho_{4.2} = f(1/T)$ on the thickness of bismuth films deposited on mica ($\operatorname{tg} \alpha$ in relative units)

of the activation energy (emphasized density of states at a certain level). The "activation energy" thus determined is, naturally, an oscillating function of the film thickness. As a result of the energy dependence of the relaxation time the redistribution of the electrons between the size subbands connected with the non-monotonous behaviour of function $g(d)$ becomes important.

The numerical calculation of the temperature dependence of the conductivity of a semi-metal film (performed by Sandomirskii, Ogrin, and Kovalev [17]), taking into account peculiar features of the energy spectrum of a thin film and assuming carrier scattering by randomly distributed impurities with δ -type

potential, agrees well with the experimental results.

The peculiar temperature dependence of the resistivity observed in bismuth films according to our view could not be explained without considering the quantum size effect.

5. Sample Deformation Phenomena

It is well known that a deformation of bulk bismuth samples is the cause of a change in the value of the overlapping of the bands [18]. In the case of thin films for which the conditions of size quantization are fulfilled, a variation in the value of the overlapping of the bands should produce a non-monotonous dependence of the kinetic coefficients on the deformation parameter.⁵⁾ The above-said non-monotony of the dependence is connected with an abrupt change in the density of electron states on the Fermi surface taking place as the overlapping value of the bands varies.

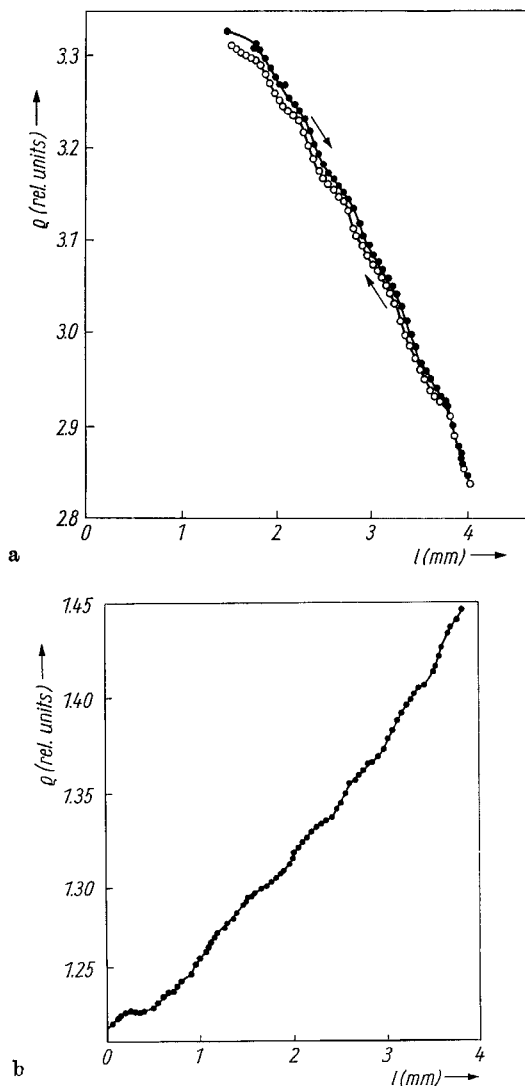
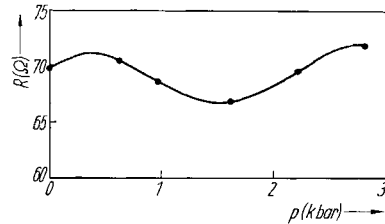


Fig. 6. a) Dependence of the resistivity of a bismuth film ($d = 1900 \text{ \AA}$) on mica on sag (compression, $T = 78^\circ\text{K}$) b) Dependence of the resistivity of a bismuth film ($d = 1900 \text{ \AA}$) on mica on sag (tension, $T = 78^\circ\text{K}$). a) and b) refer to different samples

⁵⁾ This was first suggested by Prof. S. G. Kalashnikov and later mentioned in [19].

Fig. 7. Dependence of the resistivity of a bismuth film on mica on pressure at uniform compression ($d = 2600 \text{ \AA}$, $T = 78^\circ\text{K}$)



The presence of the quantum size effect in phenomena connected with the deformation of samples was tested on bismuth films condensed on mica according to conditions that have been described above [20, 21]. The resistivity of thin layers of bismuth was measured during their sagging deformation [20] and also the dependence of the kinetic coefficients of the samples on pressure under the conditions of hydrostatic compression [21] (78 and 300 °K).

Under the conditions of sagging deformation in bismuth films (on mica) a non-monotonous decrease of the resistance under compression (Fig. 6a) and a non-monotonous increase of the resistance under tension (Fig. 6b) have been observed (the maximum value of deflection $l = 5 \text{ mm}$ corresponds to a pressure $P \approx 10^3$ to 10^4 kp/cm^2). In the region of 5000 to 6000 $\text{\AA}$ the relative value of the oscillation amplitude falls by several times; in the region $d > 20000 \text{ \AA}$ oscillations are not observed. The period of oscillations Δl depends slightly on the film thickness. One may suggest that the presence of oscillations $\rho(l)$ is connected with the quasidiscrete nature of the energy spectrum in the samples investigated.

The type of the variations of the kinetic coefficients under the conditions of hydrostatic compression [21] confirms the connection of the deformational characteristics of the samples with the quantum size effect. The dependence of the resistivity of the films on the pressure has a clearly expressed oscillatory character (Fig. 7). The period of oscillations decreases with increasing film thickness. Numerical evaluations of the period of the oscillations ΔP based on the known dependence of the Fermi energy on the pressure under uniform compression of massive Bi samples, correlate with experimentally observed $\Delta P(d)$ values.

A rise of temperature up to 300 °K leads to a decrease in the oscillation amplitude. On bulk samples the oscillations are not observed.

The above given experimental data are in a good agreement with the idea of quantum size effect.

6. Optical Phenomena

The quasi-discrete nature of the energy spectrum in thin films and the peculiar features of the density of electron states function connected therewith are reflected in a number of optical effects.

The presence of a zero energy of carriers $E_0 \approx \pi^2 \hbar^2 / 2 m^* d^2$ leads to an increase in the width of the forbidden gap (E_g) of semiconductor films with decreasing thickness [4]. This effect should result in a shift of the red edge of the optical absorption ($\hbar \nu_{\text{red}}$) with varying film thickness. The spectral dependence of the absorption coefficient in accordance with the non-monotonous

behaviour of the dependence $g(E)$ should also be of a non-monotonous nature [4].

The above-mentioned effects were investigated in bismuth films obtained by means of vacuum evaporation on a KBr substrate [22]. (The crystal are oriented with their binary axis perpendicular to the film plane.) In samples with given orientation sharply expressed oscillations have been observed of the thickness dependence of the kinetic and galvanomagnetic coefficients showing the presence of size quantization.

The spectral dependence of the optical transmission was studied in films 200 to 3000 Å thick in a wavelength region of 0.75 to 25 μm at a temperature of 300 °K.

The experimental results may be summed up as follows: An increase in energy corresponding to the absorption edge ($h\nu_{\text{red}}$) is observed, with decreasing film thickness (Fig. 8). From a value $h\nu_{\text{red}} \approx 0.07$ eV for $d \approx 3000$ Å the energy of the absorption edge increases up to 0.14 eV for $d = 300$ Å.

With further decrease in thickness to ≈ 200 Å an abrupt increase in the edge energy up to a value $h\nu_{\text{red}} \approx 0.45$ eV is observed.

The value $h\nu_{\text{red}} \approx 0.07$ eV observed on thick films corresponds to the optical width of a forbidden band in bulk bismuth (without regarding the temperature dependence of E_g).

In the case of an imperfect sample, where it proves to be difficult to obtain the quantum size effect, no shift in the absorption edge in the same thickness range is observed. The direct measurements of $n(d)$ show that the observed shift of $h\nu_{\text{red}}$ is not connected with an increase in the carrier concentration.

For about 30% of the samples tested, the spectral dependence has non-monotonous character. The extrema shift to the short-wavelength region of the spectrum as the film thickness decreases.

These experimental results (including a break of curve $h\nu_{\text{red}}(d^{-2})$ in the point $d = 300$ Å) can be understood on the basis of the quantum size effect [4, 8].

Alekseevskii and Vedenev [23] observed transmission oscillations of aluminium films in dependence on their thickness, when the films were exposed to monochromatic light ($\lambda = 0.63$ μm) at a temperature of 4.2 °K. An increase in temperature up to 300 °K results in a disappearance of the oscillations. The authors tend to interpret the relations observed as a manifestation of the quantum size effect in aluminium.

Filatov and Karpovich [24] observed a shift in the absorption edge with varying thickness of InSb films. This phenomenon is also interpreted as a manifestation of the quantum size effect.

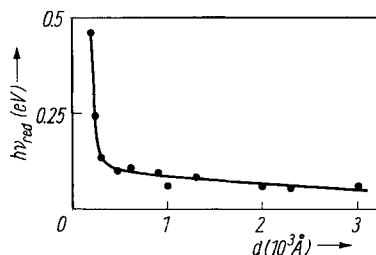


Fig. 8. Dependence of the red edge of optical transmission on the thickness of bismuth films condensed on KBr

Hence, it is possible to arrive at the conclusion that the peculiar features of the optical characteristics of thin films, which have been experimentally observed, may be considered with a great degree of probability to be induced by the quantum size effect.

7. On the Metal-to-Semiconductor Transition

Before discussing further experimental results, let us briefly consider an effect specific for semi-metals, namely, the disappearance of the overlapping of bands, the appearance of an energy gap in the spectrum of quasi-particles, and the corresponding transformation of the metallic properties of the sample into semiconductor ones. The appearance of the energy gap is connected with the variation in the zero energy of the particles as the size of the samples decreases, so that for a fairly thin film the summary shift of the extrema of the overlapping bands in various directions is found to be greater than the overlapping value. This idea was suggested in work [8], in which a method of detecting this effect by means of optical measurements was also proposed. Two years later, the same effect was studied from the quantitative standpoint in the paper [13].

The works done in order to observe this transition may be divided into two groups. The first group of works deals with the thickness and temperature dependences of the kinetic and galvanomagnetic coefficients; the second group represented only by a single work [22] is connected with optical investigations.

In the case of bismuth films with the trigonal axis being perpendicular to the film plane, the following phenomena are observed: A marked growth of the resistivity in the region of small thicknesses is found [1, 25, 15] ($d < 400 \text{ \AA}$, $T = 4.2^\circ$ to 300°K [1], $d < 400 \text{ \AA}$, $T = 300^\circ \text{K}$ [15], $d < 1000 \text{ \AA}$, $T = 300^\circ \text{K}$ [25]).

The authors [15] have observed a peculiar behaviour of the dependence $\varrho(d)$ in the point $d = 360 \text{ \AA}$ ($T = 300^\circ \text{K}$).

In work [25], in addition to a growth of $\varrho(d)$, also an increase in $R_H(d)$ and a change in the sign of the resistance temperature coefficient for $d < 1000 \text{ \AA}$ was observed.

The authors of the works [25] and [15] consider the facts stated above to be connected with the effect of the disappearance of the overlapping of the bands.

Besides the fact that these experimental peculiarities allow an explanation which is not connected with the appearance of a gap (especially results in [25] where the transition thickness $d = 1000 \text{ \AA}$ seems to be unduly high), a fact exists which contradicts the removal of the degeneracy at least for $d > 200 \text{ \AA}$. In this case we mean the high-frequency oscillations $\varrho(d)$ connected with the hole part of the Fermi surface (cf. Section 2) observed in bismuth films of the same orientation.

The optical characteristics of thin layers of bismuth of different orientation (the binary axis being perpendicular to the film plane, KBr substrate) were studied in work [22]. A break observed in the curve $h \nu_{\text{red}}(d^{-2})$ for $d \approx 300 \text{ \AA}$, may be connected with the removal of the degeneracy.

The appearance of the break in the curve is connected with the following fact: When a degeneracy exists, the growth of the edge energy with decreasing film

thickness is retarded owing to the fact that the Fermi energy changes slowly direct interband transitions. Thus the disappearance of the overlapping should be accompanied by a marked growth of $\hbar \nu_{\text{red}}(d)$.

A concrete calculation of the dependence $\hbar \nu_{\text{red}}(d)$ for an actual anisotropic bismuth model, given in work [26], yields results which agree well with the experimental data.

It should also be pointed out that in samples oriented with their binary axis perpendicular to the film plane, the holes possessing in this direction a mass $m_p^* \approx 0.06 m_0$ [12] are also quantized. This fact also favours the appearance of a gap. Nevertheless, the single fact of the presence of a break in the curve $\hbar \nu_{\text{red}}(d)$ cannot be used as a final prove of the realization of the metal-to-semiconductor transition.

In a recently published communication [27] concerning the observation of the considered transition on $\text{Bi}_2\text{Te}_3\text{S}_5$ films, no sufficient data are given characterizing the quality of films of abnormally small thicknesses (few atomic layers).

Hence, the experimental results available do not allow a final conclusion with regard to the actual existence of the metal-to-semiconductor transition in thin films.

8. Tunnel Effect

The method of tunnel spectroscopy may be used in order to detect the specific nature of the density of electron states in a thin film. Let us consider a tunnel system consisting of a size-quantized film and a bulk metal separated by a dielectric gap. Fig. 9 represents the energy diagram of such a system. It is seen from Fig. 9 that with varying voltage applied to the tunnel gap, the current-voltage characteristic will show peculiarities each time when the Fermi level of the massive electrode intersects the quasi-discrete level in the film. This method allows the direct probing of the density of states function and the energy spectrum of the carriers.

Problems connected with tunnel peculiarities in thin films were studied theoretically in the works [28], [6], and [9].

The tunnel effect was experimentally studied for systems of the following combinations: 1. bulk bismuth-bulk silver (thick films of a corresponding substance were used as "bulk" ones, in which obviously no size quantization occurred); 2. thin film Bi-bulk Bi; 3. thin film Bi-bulk Ag [2, 29]. Films 800 to 2000 Å thick were studied as "thin" ones.

The bismuth films were obtained on a mica substrate by using the method described in Section 1, and they possessed the same structural and electrical

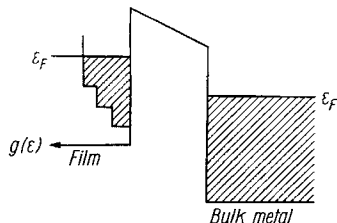


Fig. 9. Energy diagram of the tunnel system: film - dielectric - bulk metal

characteristics of the samples used for studying the galvanomagnetic and kinetic phenomena.

The authors did not use a solid dielectric as a tunnel gap in order to avoid the influence of undesirable processes in oxide layers, for example, tunneling via trap levels. An interelectrode gap was formed by superimposing two independently manufactured elements due to the presence of a corresponding difference in thicknesses.

Fig. 10 shows the configuration of the samples and a cross-section of the assembled tunnel system.

Narrow strips ($1 \times 0.1 \text{ mm}^2$) extending from the contact leaves on a lower electrode, form nine tunnel diodes in conjunction with the upper film. The contact with the upper electrode is effected by virtue of a contact projection connected with one of the contact leaves of the lower electrode. The described system is inserted in a block with push contacts, and is clamped with the aid of optically polished glasses.

Imperfections of the surface result in that three to five diodes out of nine are found to be short-circuited.

When carrying out the measurements, the system is immersed directly into liquid nitrogen (or liquid helium).

The described tunnel system was used for performing measurements of current-voltage characteristics by means of the compensation method, whereas the dependence of the differential conductivity on the voltage ($dI/dU(U)$) applied to the diode was studied by means of a differential technique, including the possibility of compensating the ohmic component of the signal.

The resistance of the tunnel junctions was usually found to be equal to 1 to 10 M Ω .

The principal results consist in the following: In the case of "bulk" electrodes Bi-Ag, the current-voltage characteristics are monotonous.

In the same voltage range (up to 100 mV) the dependence $dI/dU(U)$ shows bumps in the points +30 mV, +60 mV, +80 mV, -35 mV, and -100 mV (the sign + corresponds to a positive potential on bismuth with regard to silver), i.e. exactly in those points which, according to observations of Esaki

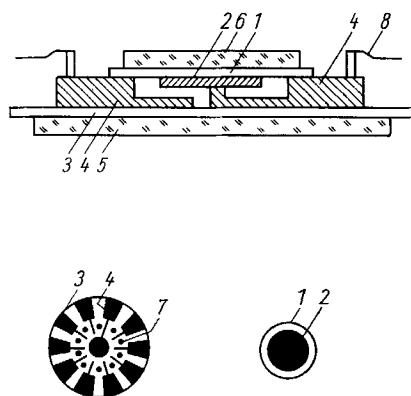


Fig. 10. Configuration of the samples and schematic diagram representing the tunnel system (1,3 — mica substrates; 2,4 — films; 5,6 — polished glasses; 7 — additional supports; 8 — push contacts)

[30], correspond to the edges of the energy bands in bulk bismuth. (These bumps are naturally imperceptible on current-voltage characteristics ($I(U)$) on account of an insufficient sensitivity of the method.)

An example of the dependence $dI/dU(U)$ for thick bismuth films is given in Fig. 11.

The main phenomenon observable in systems thin film Bi-bulk Bi (or Ag) is the presence of oscillations in the characteristics $I(U)$ and $dI/dU(U)$ (see Fig. 12 and 13, respectively).

The distance between the bumps and the amplitude of the oscillations depends on the polarity of the voltage applied. In the case of negative polarity on the thin film in a certain voltage interval a monotonous dependence is substituted for the non-monotonous one ($I(U)$) (Fig. 12).

The potential difference at which the type of the dependence changes determines, according to the energy diagram shown in Fig. 9, the position of the Fermi level in the film.

The values of E_F obtained by the above method lie in a range of 0.02 to 0.028 eV, i.e., agree well with the values of the Fermi energy in bulk samples. Furthermore, the number of peculiarities of the aforementioned characteristics is essentially a function of the film thickness (Fig. 13a) and corresponds to the number of the quasi-discrete levels located below the Fermi energy in a film of given thickness. Finally, the distance between the bumps allows the value of the component of the effective mass in the trigonal direction, m_z^* to be determined. The value agrees well with the value m_z^* known for bulk bismuth. Fig. 13b illustrates the reproducibility of the results by showing the distribution of the bumps along the voltage scale for the fixed thickness samples tested (for $d = 1000 \text{ \AA}$).

We believe that these results constitute an independent and unambiguous prove of the realization of the quantum size effect in the objects being studied. Detailed measurements of the characteristics of the tunnel effect carried out in a wide thickness range authorize us to expect that it is possible to obtain a thor-

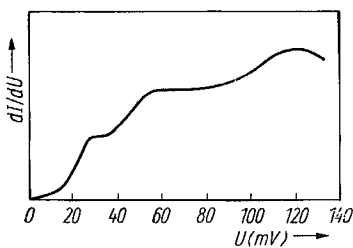


Fig. 11. Tunnel characteristic for the "bulk" bismuth (Bi-Ag system, thickness of the Bi film: 5000 Å; Bi is positively charged with respect to Ag; $T = 78^\circ \text{K}$)

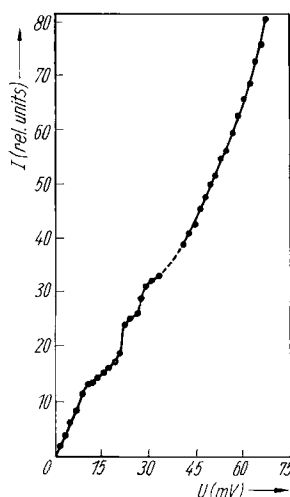


Fig. 12. Current-voltage characteristic of the tunnel system (Bi film — 1000 Å thick; the Bi film is negatively charged with respect to bulk Bi; $T = 78^\circ \text{K}$)

Fig. 13a. Tunnel characteristics dI/dU vs. U of bismuth films 1000 and 2000 Å thick (Bi-Ag system; the Bi film is charged negatively with respect to Ag; $T = 78^\circ\text{K}$)

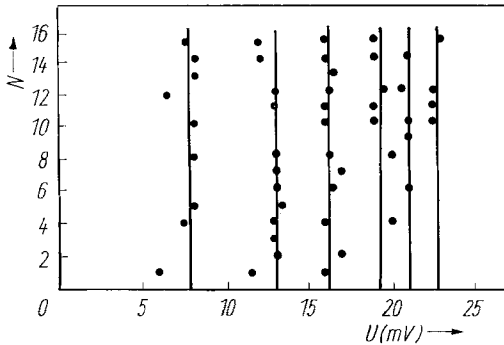
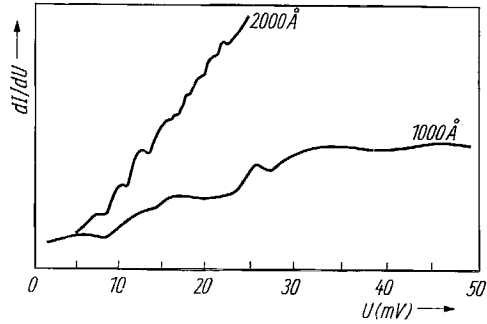


Fig. 13b. Distribution of the bumps along the voltage scale for various samples 1000 Å thick (number of samples being plotted on the ordinate axis)

ough representation of the change of the energy spectrum of electrons when passing over from a bulk sample to a thin film.

9. Quantizing Magnetic Field Phenomena

When combining the size quantization with the quantization in a magnetic field qualitatively quite new effects appear.

We shall consider some phenomena taking place in a thin film placed into a strong magnetic field oriented perpendicular to the film plane (according to the z -axis). In this case, the energy spectrum of the carriers undergoes a radical remodelling. The energy spectrum becomes completely discrete, i.e., the energy E is essentially a function of only discrete quantum numbers:

$$E = \hbar \omega_c \left(m + \frac{1}{2} \right) + \frac{\pi^2 \hbar^2}{2 m^* d^2} n^2,$$

where ω_c is the cyclotron frequency, $m = 0, 1, 2, 3, \dots$ is the magnetic quantum number (number of the Landau band), and $n = 1, 2, 3, \dots$ is the size quantum number (number of the size subband).

In coordinates $E(|K_z|)$ ($K_z = (\pi/d)n$) the given spectrum appears as a number of separate points arranged along the corresponding Landau band (Fig. 14).

The combination of the size quantization with the quantization in a transverse magnetic field yields new possibilities for investigating the structure of the electron energy spectrum [31].

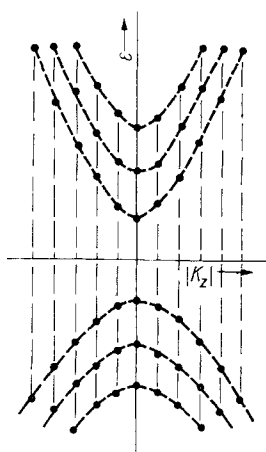


Fig. 14. Discrete energy spectrum of carriers in a thin film placed into a transverse quantizing magnetic field

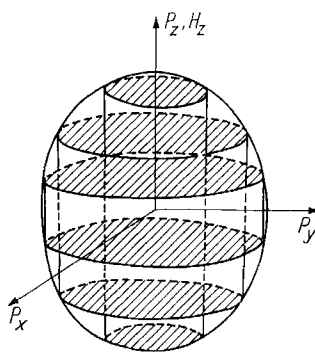


Fig. 15. Allowed cross-sections of the Fermi surface in a thin film

First of all, in conformity with the discrete energy spectrum, a striped optical absorption spectrum should be observed. The measurement of the position of the absorption lines allows, in principle, to reconstruct the shape of the Landau bands corresponding to the various values of the magnetic quantum number.

For an analysis of the absorption spectra it is necessary to take the selection rules into account.

For a model of a semiconductor film with a standard band scheme placed into a magnetic field, the selection rules for direct optical interband transitions are found to be: $m^{(1)} = m^{(2)}$, $n^{(1)} = n^{(2)}$, $K_y^{(1)} = K_y^{(2)}$, where the indices in the brackets characterize: (1) — the valence band, (2) — the conduction band; K_y — the y -component of the wave vector of electrons. The vector potential of the magnetic field is assumed as $A_x = A_z = 0$, $A_y = -Hx$.

Moreover, the study of oscillatory effects is of substantial interest. It is well known [32] that in bulk samples the period of oscillations of the thermodynamic values of a degenerate Fermi gas with varying magnetic field is determined by the area of the extreme cross-sections of the Fermi surface. The non-extreme sections are connected with the monotonous part of the thermodynamic potential, and the areas of these cross-sections cannot be determined by investigating the effects of the same type as the de-Haas-van Alphen effect.

In a thin film the situation is essentially changed. The number of sections of the Fermi surface allowed by means of the size quantization may be rather small (Fig. 15). In this case, each of the allowed sections acts as an extreme one, i.e. gives a contribution to the oscillatory part of the thermodynamic potential in the form of harmonics whose frequency is determined by the area of the cross-section under consideration.

The relation between the oscillation period and the cross-sectional area of the corresponding non-extreme section may be determined as follows [31]:

The thermodynamic potential Ω of a thin film placed in a transverse magnetic field may be expressed as

$$\Omega = -kT \frac{L_x L_y}{\pi \hbar c} e H \sum_m \sum_n \ln \left(1 + \exp \frac{E_F - E_{m,n}}{kT} \right), \quad (1)$$

where L_x, L_y are the dimensions of the sample taken in the film plane.

The energy spectrum of a film, approximated by the model of a potential channel with infinitely high walls and a flat bottom, is determined by the quasi-classical condition for quantization:

$$S(E, P_z) = (m + \gamma) \frac{2 \pi e \hbar H}{c},$$

where $S(E, P_z)$ is the cross-section area of the iso-energetic surface intersected by the plane $P_z = (\pi \hbar/d) n^6$; γ is a number varying between zero and 1.

Supposing large values of m , the stated condition for quantization does not impose limitations on the size quantum number n . Using small values of n it is possible to determine the areas of the non-extreme sections.

When summing up in expression (1) with respect to the index m with the aid of Poisson's formula and employing only the first harmonic, we obtain the period of the oscillations of the thermodynamic values connected with an allowed section for varying magnetic field and fixed film thickness as,

$$\Delta \left(\frac{1}{H} \right)_n = \frac{2 \pi e \hbar}{c S_n(E_F)},$$

where $S_n(E_F)$ is the area of an allowed cross-section of the Fermi surface corresponding to the quantum number n .

Proceeding in the same manner, we can obtain an expression for the period of oscillations of the thermodynamic values in the case where the film thickness is varying and the value of the magnetic field is fixed. In this case, the summing up is made with respect to the index n . The possible minimum values of n are thereby determined by the conditions of suitability of the Poisson's formula.

The period Δd_m is expressed as follows:

$$\Delta d_m = \frac{\pi \hbar}{P_z^{(m)}},$$

where $P_z^{(m)}$ is the z -component of the Fermi momentum corresponding to a magnetic subband with a number m (half height of the cylinders in Fig. 15).

Consequently, the measurements of the oscillation characteristics in each of the cases under consideration (fixed thickness or fixed magnetic field) as well as their combination permit in principle to reconstruct the Fermi surface.

The conclusions made here were confirmed experimentally [33, 34]. In the investigations bismuth films deposited on mica were used, for which the presence of the size quantization was previously established by measuring the dependence $\varrho(d)$.

The differential magnetoresistance $\partial \varrho / \partial H(H)$ as well as the dependences of $\partial^2 \varrho / \partial H^2$ on the magnetic field directed perpendicular to the film plane (along the trigonal crystal axis) are determined. Measurements were carried out on films 800 to 3000 Å thick at 4.2 °K with the aid of a standard differential technique in the process of charging or discharging a superconducting solenoid.

⁶⁾ It is suggested that the symmetry plane of the Fermi surface is parallel to the film plane.

The main experimental results consist in the following: First of all, oscillatory dependences $\partial\rho/\partial H(H)$ and $\partial^2\rho/\partial H^2(H)$ (Fig. 16 and 17) are observed whose period varies from 10^{-3} to 10^{-4} G $^{-1}$. In a bulk sample the period corresponding to the area of an electron extreme cross-section, with the same orientation of the magnetic field, equals to $\approx 1.2 \times 10^{-5}$ G $^{-1}$ [35]. The high frequency oscillations corresponding to cross-section near the extremal one are also observed at curves $\partial^2\rho/\partial H^2(H)$.

It is reasonable to conclude that the low-frequency oscillations of such a great period (which is not observed in bulk samples and in imperfect non-quantized films) are connected with small non-extreme cross-sections in the electron part of the Fermi surface. This conclusion is confirmed by the characteristic dependence of the oscillation frequency $1/\Delta(H^{-1})$ on the film thickness d (Fig. 18). The figure shows a decrease in the cross-section area and a subsequent "eliminating" of separate sections from the volume limited by the Fermi surface (Fig. 15) as the film thickness decreases. The "eliminated" section is gradually replaced by the following allowed section with smaller quantum number n . It is also apparent that at a certain thickness (on the second derivative) two periods corresponding to two allowed sections are simultaneously observed: Three non-extreme sections are observable for a thickness ≈ 1020 Å. It is important to point out that the period of the "falling-out" sections determined by the points in which the frequency $1/\Delta(H^{-1})$ becomes equal to zero (Fig. 18) coincides with the period of oscillations of the dependence $\rho(d)$ obtained for the same samples.

The data presented in Fig. 18 allow a study of the topology of the Fermi surface to be made.

In addition to the oscillatory dependences $\partial\rho/\partial H(H)$ with a large (according to $1/H$) period, $\partial\rho/\partial H$ curves with one or two bumps (Fig. 16, curves (b)) and smooth curves (Fig. 16, curve (c)) are also observed. Let us also point out that the low-frequency oscillations are detected in relatively weak fields (500 to 5000 G), for

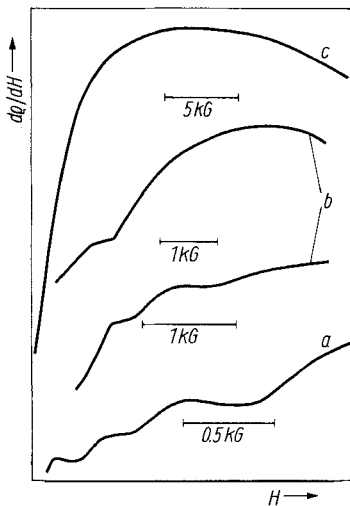


Fig. 16. Dependence of the differential magnetoresistance on the magnetic field of bismuth films deposited on mica. (a) $d \approx 1200$ Å, (b) $d \approx 1100$ and 900 Å, (c) $d \approx 1700$ Å; $T = 4.2$ °K

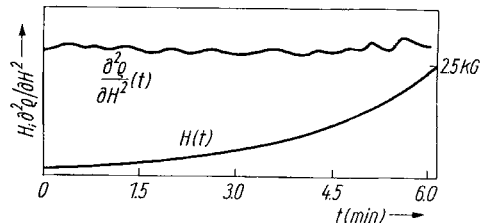


Fig. 17. Example for the dependence $\partial^2\rho/\partial H^2(t)$ and $H(t)$ for a bismuth film on mica ($d = 1000$ Å)

Fig. 18. Dependence of the oscillation frequency $1/\Delta(H^{-1})$ on the thickness of bismuth films on mica and $g(d)$ of the same films

which the criterion of a strong field $\mu H > 1$ is not fulfilled ($\mu \approx 20000 \text{ cm}^2/\text{Vs}$ for $d \approx 2000 \text{ \AA}$). These facts may be explained as follows:

The bismuth films are essentially a mosaic single crystal with average crystallite size equal to 3 to 5 μm . It is reasonable to suppose that the scattering of carriers occurs mainly on grain boundaries. In this case, the quantization conditions in a magnetic field will be determined by the ratio between the crystallite sizes and diameter of the electron orbit. As the shape of the orbit in the geometrical space is like that of an orbit in momentum space, the quantization conditions in the magnetic field for electrons pertaining to a small allowed section are realized for weaker fields than those for electrons corresponding to a large section area.

Curves (b) shown in Fig. 16 correspond to the case when within of one size subband referred to a small section, only one or two Landau levels are located below the Fermi level.

Finally, curve (c) of Fig. 16 is characterized by such a film thickness that the filled small section is absent, whereas the larger area section is imperceptible on account of an unfavourable ratio between the diameter of the orbit and the size of the crystallites.

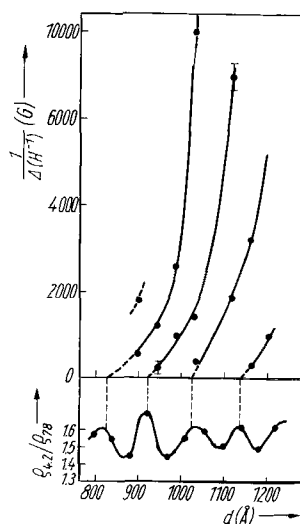
Amongst the phenomena considered one possibility of tunnel spectroscopy in a strong magnetic field should be discussed now. If a system composed of two thin films separated by a dielectric gap is placed into a transverse magnetic field, the energy spectrum of the current carriers on both sides of the dielectric will be essentially a set of discrete levels. In such a system a tunnel current is possible only at voltages corresponding to the coincidence of levels located on the left and right of a potential barrier. Hence, the current-voltage characteristic of such a system will be represented by separate current peaks with corresponding values of the voltage determined by the nature of the energy spectrum of current carriers in films, being changed by varying the magnetic field and thickness of the specimens.

A number of interesting effects observable in a thin film placed into a magnetic field are theoretically studied in the works of Tavger and Eruchimov [36].

10. Superconductivity

The presence of a non-monotonous dependence of the function $g(d)$ should result, according to [37], in a new effect in thin superconducting films. With varying film thickness the superconductivity gap and critical temperature (T_c) should oscillate with a period equal to half of the de Broglie wavelength and an attenuating amplitude.

Komnik and Bukhshtab [38] have attempted to observe the Blatt-Thompson effect on tin films. Tin samples of varying thickness (condensed on a glass



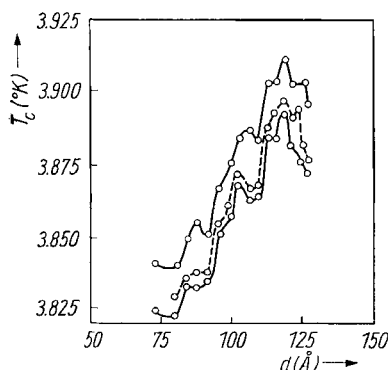


Fig. 19. Dependence of the critical temperature on the thickness of superconducting thin films of tin (see work [38])

substrate cooled to a temperature of 200 °K) were found to possess a distinct texture. Direction [100] coincides with the normal to the film.

The dependence $T_c(d)$ was actually found to be non-monotonous, where according to the observations of the authors [38] oscillations of two types are detected, namely with periods 15 Å and ≈ 7 Å (Fig. 19).

The oscillatory dependence of the conductivity on the film thickness with a period ≈ 7 Å was observed on the same samples. The above-said period well corresponds to the known value of the Fermi quasi-momentum in the [100] direction.

These facts in spite of the very small amplitude of the oscillations (connected with the structural imperfection of the samples) and difficulties encountered when determining the small period, apparently show the realization of the quantum size effect in the tin films.

11. Conclusions

The experimental results stated above make it possible to establish unambiguously that the quantum size effect is actually realized.

The experimental investigations thus carried out, which in fact are only an initial stage of investigating this new phenomenon, have already shown additional possibilities for studying the structure of an electron spectrum by means of the quantum size effect. It is quite possible to assume that these possibilities are not limited for studying the structure of an electron spectrum, but can also be applied for investigating the spectrum of phonons and some other fundamental characteristics of solids.

Finally, these investigations have clarified some aspects of the practical use of the quantum size effect. In principle, thin films may be employed as radiation detectors with an adjustable wavelength range. In resonant tunnel systems, it is possible to obtain current-voltage characteristics with several sections of negative resistance [6]. The quantum size effect taken in conjunction with the quantization in a magnetic field may in principle be employed for the construction of an injection laser [39].

Acknowledgements

The author expresses his sincere gratitude to Prof. Dr. M. I. Elinson, E. P. Fesenko, Dr. Yu. F. Ogrin, D. N. Korneev, L. A. Kulik, R. N. Sheftal, Dr. V. B. Sandomirskii, and Dr. T. N. Pinsker for their help in discussing the problems raised in the present paper.

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(Received December 10, 1969)