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QUANTUM SIZE EFFECTS IN SEMICONDUCTING AND SEMIMETALLIC FILMS

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I. INTRODUCTION. CONDITIONS FOR THE EXISTENCE OF QUANTUM SIZE EFFECTS IN FILMS

INTEREST in the physical properties of films has increased recently. This is connected primarily with the needs of the rapidly developing field of microelectronics. The investigation of films is of interest both from the purely scientific point of view and from the practical one, since new specific effects, not observed in bulky samples, can appear in the film state. In addition, the investigation of crystalline films yields definite information concerning the physical properties of the crystal.

The investigation of solids in different practical devices frequently imposes definite requirements on the energy spectrum of the carriers. By subjecting the sample to the action of strong magnetic or electric fields, to high pressure, by changing the character of the doping, etc., it is possible to obtain crystals with the required characteristics, in particular, with a designated energy spectrum. The shape and the dimensions of the sample also influence the physical properties of the crystal. This influence will become significant when the dimensions of the sample are of the order of some characteristic length. In particular, when the dimensions of the crystal sample are comparable with the electron mean free path, the kinetic properties, such as the electric conductivity, become dependent on the dimensions and shape of the crystal.

Variations can be expected in the physical properties of crystals also in the case when the finite dimensions of the sample lead to quantization of the quasiparticle motion. This was pointed out back in the 30's[1].

The idea of the size quantization found its reflection in the theory of oscillations of thermodynamic characteristics of thin layers of metal, developed by I. M. Lifshitz and his co-workers[2,3] (a complete review and a bibliography of this cycle of investigations are given in[4]). Size quantization is also important in finely-dispersed media[5,6].

Recently, interest in the study of the quantization of quasiparticles in films has increased in connection with the extensive utilization of thin semiconducting films in microelectronics.

In metals, the wavelength is on the order of the lattice period a. Therefore, in actual metallic films containing many atomic layers, quantum size effects are weakly pronounced and have the character of corrections to the properties of bulky samples. The situation is different in semiconducting and semimetallic films, since the de Broglie wavelength of the carriers in semiconductors and semimetals can be larger by several orders of magnitude than the interatomic distances. The de Broglie wavelength depends in this case on the effective carrier mass m^*, their concentration n, and temperature T. Therefore, under definite conditions, which

are discussed below, size quantization in semiconducting and semimetallic films becomes already significant at thicknesses $L \sim 10^{-5}$ cm (see below) and leads to qualitatively new effects not observed in bulky samples.

A film is an example of a bounded medium. This circumstance leaves a significant imprint on the properties of the quasiparticles in films. Owing to the limited size of the film in one direction, the projection of the quasi-momentum k_z perpendicular to the plane of the film is indeterminate. Therefore, the energy of the electron in the film, as well as the energy of any other quasiparticle, is determined by the longitudinal projections of the quasimomentum k_x, k_y and, generally speaking, by a discrete quantum number s, which replaces k_z:
$\mathscr{E} = \mathscr{E}(k_z, k_y, s)$. At a fixed s, the energy runs continuously through an interval of values which will be called henceforth subband. The longitudinal projections of the quasimomentum are determined in a two-dimensional Brillouin zone, which in the case of a film, generally speaking, is not a plane cross section of a three-dimensional band (see Chapter II). The surface states are also defined in terms of a quasimomentum from a two-dimensional Brillouin zone, and the energy values corresponding to these states also form one subband. The subbands usually overlap, since the energy interval $\mathscr{E}_{s+1} - \mathscr{E}_s$ for fixed k_x and k_y is as a rule smaller than the width of the subband. Thus, the energy spectrum of the electron in the film is quasidiscrete and consists of overlapping subbands.

Quantization of the transverse motion in films can appear, however, only if a number of conditions are satisfied[7,8]. Owing to the scattering of the electrons, the quasidiscrete spectrum is partially smeared out. In order for the quasidiscrete character of the spectrum to be maintained, it is necessary that the smearing h/τ (τ—relaxation time) be smaller than the distance between the neighboring subbands:

$$\frac{h}{\tau} \ll \mathscr{E}_{s+1} - \mathscr{E}_s. \tag{1.1}$$

For an exact determination of τ it would be necessary to calculate the damping of the quasiparticles in the film for different possible scattering mechanisms, as was done for example, by Rytova for the electron-electron and electron-impurity scatterings[9]. Since it is impossible to obtain a sufficiently accurate theoretical estimate, it is necessary to determine the relaxation time experimentally. It is convenient, for example, for estimating purposes, to express it in terms of the experimentally measured mobility. This means that the transport relaxation time, which takes into account all the carrier-scattering mechanisms in the film, is substituted in (1.1).

In addition to the condition imposed on the relaxation time (or mobility), there should also exist a limitation

on the temperature and concentration of the carriers, in order that the number of populated subbands be small. In this case the difference between the properties of the film and the properties of the bulky sample is more clearly pronounced. The condition for the concentration follows already from dimensionality considerations. In order for only one subband to be populated, it is necessary to satisfy the inequality

$$n < \frac{A}{L^3} \; ; \tag{1.2}$$

A—dimensionless coefficient that depends on the quantization law.

Upon quantization we get

$$\mathcal{E}_s = \frac{\hbar^2}{2m^*} \left(\frac{\pi}{L} \right)^2 s^2$$

(the film is approximated by an infinitely deep well, m*—effective mass in the direction perpendicular to the film), A = $3\pi/2$.

The relation for the temperature is determined from the requirement that the thermal spread be small compared with the distance between subbands:

$$k_B T < \mathcal{E}_{s+1} - \mathcal{E}_s. \tag{1.3}$$

It should also be noted that in order to observe the quantization effects the film should be sufficiently homogeneous in thickness[10]. It can be shown that the quasidiscrete character of the spectrum, starting with the s-th subband, will not be violated if the relative spreading of the thickness is smaller than $\Delta L/L$. In real semiconducting and semimetallic films, the conditions imposed on the concentration, temperature, and thickness homogeneity can be relatively easily satisfied for thicknesses L $\lesssim 10^{-5}$ cm. The relation (1.3) for an effective mass of $0.01 m_0$ is satisfied already at room temperatures. As seen from (1.2), for the same thickness required to populate one subband, the concentration should be on the order of 10^{16} cm^{-3}. Thus, at concentrations of 10^{17} cm^{-3}, which can be realized in semimetallic and semiconducting films, only a few subbands are populated; as to metals, by virtue of the large carrier concentration, a large number of subbands is populated for thicknesses up to L $\sim 10^{-7}$ cm, thus decreasing the influence of quantization. The most stringent requirement for the existence of a quasidiscrete spectrum in a film is condition (1.1), which is satisfied only in sufficiently perfect and pure films. To satisfy condition (1.1), relatively high mobilities are required. For example, at a thickness L $\sim 5 \times 10^{-6}$ cm, the mobility μ should be larger than 10^3 cm^2/V-sec. This requirement also limits the maximum temperature since μ as a rule decreases with increasing temperature. A possible exception is the case when the scattering by an ionized impurity is significant.

Quantum size effects in semiconducting and semimetallic films are not fully identical. In semimetallic films, as a rule, the current carriers are degenerate. Therefore, oscillatory effects (see below) are well pronounced in semimetals. In semiconductors, on the other hand, in order to observe oscillatory effects a high doping level is necessary, leading to a decreased carrier mobility. However, the large variety of semiconducting materials makes it possible to choose a film with better parameters than in the case of a semimetal.

Until recently, there was no convincing experimental proof of the existence of quantization of the transverse motion of electrons in films. To be sure, Crittenden and Hoffman[11] observed certain deviations of the resistance in thin films of nickel from the values predicted by the classical theory. These authors were inclined to ascribe these deviations to the influence of electron-energy quantization. They assumed that the presence of quantization leads to a decrease in the number of possible transitions, since only discrete values of the transverse momentum are allowed in the film. This, in the opinion of the authors of[11], leads to a decrease in the resistance (compared with the classical theory) with decreasing film thickness. However, this explanation of the peculiarities of the experiment is untrue, since the quantization (see Sec. 3) cannot lead to a monotonic decrease of the resistance with decreasing thickness. The first convincing experimental confirmation of the presence of a quasidiscrete electron spectrum was obtained in thin semimetallic films. Ogrin et al.[12] observed in Bi films oscillations of the mobility, of the Hall constant, and of the magnetoresistance with variation of the film thickness (see Fig. 5 below). The films investigated in[12], which were obtained by vaporization in vacuum onto mica, were of sufficiently perfect crystalline structure, and the mobility in them exceeded 10^3 cm^2/V-sec. The results of[12] are confirmed by more detailed investigations carried out in a wider thickness interval[13] L $\sim 200-5000$ Å, and in the temperature interval T $\sim 4-200°$K[14]. Similar results were obtained by Komnik[15], who investigated Bi films vaporized on a glass substrate.

A direct observation of the quantization of the motion of electrons was carried out by Lutskiĭ et al.[16] in experiments on the tunneling of electrons between thin films through a gap.

Such a method of experimentally studying the energy spectrum was proposed* by the authors of[10] (see Sec. 2, Item 4).

The quantization of the transverse electron motion was observed by Komnik and Bukhshtab in another semimetal—antimony[17]. They observed resistance oscillations with varying thickness in the range L $\sim 100-400$ Å. Instead of using a set of films of different thickness, they used in[17] a sample in the form of a wedge, thus increasing the accuracy with which the film thickness was determined.

It should be noted here that effects analogous to quantum size effects in films can occur in the thin surface layer of a bulky sample in the case when the energy bands are sufficiently deflected. Just as in a film, the deflection of the bands limits the carrier motion perpendicular to the sample surface. As a result, when a number of conditions are satisfied, the carrier spectrum becomes quasidiscrete $\mathcal{E} = \mathcal{E}$ (k_x, k_y, s). In the case of a many-valley single crystal, just as in a film[18], level splitting is possible and can lead to a decrease in the number of equivalent valleys.

The surface deflection of the bands can be varied by means of an external electric field applied perpendicularly to the sample surface. In some sense, a change in the magnitude of the field is analogous to a change in the film thickness, and can lead to similar effects. A discussion of the influence of quantization on the field effect was apparently first presented by Schrieffer in[19]. By now, an appreciable number of papers devoted to quantization in the surface layer have been published[20,21]. The most detailed experimental investigation is reported in[21], where oscillations of the magnetoresistance were observed in the surface layer in a quantizing magnetic field. In this investigation they observed the case of population of one subband. So far, however, the case of filling of several subbands was not realized under experimental conditions. A detailed discussion of the quantization effect in the surface layer is beyond the scope of the present review.

The field effect can also be used in principle to study the properties of films with a quantized spectrum. This method, proposed by Sandomirskiĭ[22], consists in the following. The investigated film serves as one of the plates of a capacitor. By varying the potential difference it is possible to vary the electron concentration in the film in such a way, that the number of filled subbands changes. This can lead to an oscillatory dependence of the mobility and the conductivity on the voltage applied to the capacitor. We note that the carrier concentration in the film can also be varied by subjecting the film to pressure.

An experimental study of this effect on Bi films[23] has shown that the oscillations actually take place in the thickness interval $L \sim 2000-3000$ Å. However, it is impossible to present an exact interpretation of the obtained results by assuming that the transverse field leads only to a change of the concentration. At small thicknesses, apparently, the penetration of the field in the film becomes important, and this should lead to a change of the energy spectrum in the film. This circumstance makes an analysis of the experiment on the field effect in the film very difficult.

The number of quantum size effects is not confined to the specific dependence of the kinetic and thermodynamic characteristics on the film thickness. Quantization of the motion should also lead to a change in the temperature dependence of the film properties, to a new dependence of these properties on the magnitude and direction of the magnetic and electric fields; the quasi-discrete character of the spectrum leads to different resonance phenomena when electromagnetic radiation[24,25], sound, and electron current, etc. pass through the film.

Effects connected with the quantization of motion of other quasiparticles (magnons, excitons, etc.) in the film have been investigated in a number of experimental and theoretical studies. The present review is devoted to an analysis of effects due only to the quantization of electron (and hole) motion.

II. FEATURES OF ELECTRON STATES IN THIN FILMS

A quantitative calculation of the energy spectrum and of the wave functions in films is extremely difficult. The main difficulties are connected both with the complicated nature of the boundary conditions and with the indeterminacy of the self-consistent potential in the film, which has a lower symmetry than in the bulky sample. Certain special models and approximations used by different authors (see[26]) for the calculation of the spectrum cannot serve for a quantitative calculation of the physical parameters of the films. At the same time, the limitation of the electron motion in the film in one dimension leads to a number of specific features, regardless of the concrete boundary conditions and of the form of the potential inside the film.

1. Energy Spectrum in Films

The electrons in the film move in a crystal field having not a three-dimensional but only a two-dimensional translational symmetry. Even this alone leads to certain conclusions concerning the character of the electronic states in the film. The wave function will have the Bloch form

$$\psi_k(\mathbf{r}) = e^{i\mathbf{k}\mathbf{r}} u_k(\mathbf{r}),$$

where $u_k(\mathbf{r})$ has a planar translational symmetry, and k is the quasimomentum from the two-dimensional Brillouin zone. When k varies within the limits of the two-dimensional Brillouin zone, the energy runs through a continuous set of values, forming a subband. As noted in Chapter I, these planar subbands, generally speaking, are strongly overlapping.

The planar Brillouin band is determined by the two-dimensional translational symmetry of the film structure. In the case of germanium and silicon crystals, when the film is grown in the [111] direction, the structure of the film has the translational symmetry of a planar hexagonal lattice. In this case, therefore, the Brillouin zone is a regular hexagon. A similar Brillouin zone is produced in bismuth films grown along the trigonal axis (it is precisely in such films in which quantum size effects were observed in[12-16,23]).

The lowering of the symmetry on going from a bulky crystal to the film should lead to a partial or complete lifting of the degeneracy, which can arise with the merging of the bands at singular points of a three-dimensional k-space, or else with the many-valley character, when several points in the Brillouin zone correspond to one extremum. In the latter case, the lowering of the symmetry should lead to a decrease in the number of equivalent valleys. On the basis of a group-theoretical analysis it is possible to clarify the character of the energy-level splitting near the extremal points[18]. For example, in Ge and Si films, the degeneracy of the edge of the valence band, which corresponds to a quasi-momentum k = 0, is completely lifted. In the conduction band of germanium, the four-valley energy minimum splits into a three-valley minimum and a single-valley minimum. The single-valley minimum lies in the center of the two-dimensional Brillouin zone, and the points corresponding to the position of the three-valley minimum lie at the midpoints of the hexagon forming the two-dimensional Brillouin zone (Fig. 1). Generally speaking, the energy minimum in the film is shifted upward as compared with the minimum in the bulky crystal, by an amount on the order of $h^2\pi^2/2m^*L^2$. This follows from the uncertainty principle.

For the single-valley minimum in the Ge film, this shift will be smaller, since it is determined by the longitudinal effective mass, which is maximal. Unlike Ge films, the splitting of the six-valley minimum of the conduction band does not take place in Si films grown in the [111] direction.

Of great interest is the study of the spectrum in thin Bi films, in connection with the fact that it is precisely in these films that quantization was observed. As is well known, the semimetallic properties of Bi are due to the intersection of two energy bands. As a result, carriers of both polarities appear, with a concentration on the order of 10^{17}cm^{-3}. The Fermi surface of the electrons is made-up of three ellipsoids, which go over into one another when rotated 120° with respect to the trigonal axis. The usual growth direction of the crystal bismuth film is the direction of the trigonal axis. The effective mass in this direction is 0.01 m_0, and the value of the effective mass of the holes is 0.7 m_0[55].

On going over to the film, the lowering of the symmetry does not lead to a splitting of the three-valley minimum. In the two-dimensional Brillouin zone, which, as already noted, is a regular hexagon, the points corresponding to the three-valley electron minimum are the midpoints of the sides of the hexagon.

2. Density of States

To find the thermodynamic and kinetic coefficients in thin films, it is necessary to determine the density of the electronic states $G(\mathscr{E}) = d\mathscr{x}/d\mathscr{E}$, which must depend strongly on the film thickness.

The density of the states is determined by the dispersion law. Since the quasimomentum of the electron along the film is determined, the dispersion law has in the effective-mass approximation the form

$$\mathscr{E}_{k_x, k_y, s} = \frac{\hbar^2}{2m_1}(k_x - k_x^0)^2 + \frac{\hbar^2}{2m_2}(k_y - k_y^0)^2 + \mathscr{E}_s \qquad (2.1)$$

(k_x^0 and k_y^0 are directed in this case parallel to the axes of the ellipsoid). As shown in[18], as a rule there is no degeneracy near the extremal points in films. Therefore formula (2.1) is sufficiently general for semiconducting and semimetallic films. Using (2.1), we readily obtain for one subband

$$G_{sub}(\mathscr{E}) = \frac{r\sqrt{m_1 m_2}}{\pi \hbar^2} S, \qquad (2.2)$$

where r—number of two-dimensional valleys, S—area of the film. For comparison with the density of states in the bulky crystal, it is necessary to introduce the state density per unit volume of the film:

$$N_{sub} = \frac{G_{sub}}{V} = r\frac{\sqrt{m_1 m_2}}{\pi \hbar^2}\frac{1}{L}. \qquad (2.3)$$

Thus, the state density in the subband does not depend on the energy and on the number of the subband, and is inversely proportional to the thickness.

For the surface levels, the density of states is also proportional to 1/L, since the levels form a two-dimensional subband. The energy dependence, generally speaking, should be determined by the concrete dispersion law. On the other hand, near the edges of the surface subband, where the dispersion law can be regarded as quadratic, the density of states does not depend on the energy.
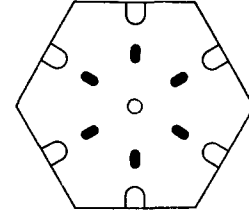

FIG. 1

The total state density N_f in the film at a given energy is equal to the sum of N_{sub} over all the subbands for which $\mathscr{E}_s < \mathscr{E}$:

$$N_f = m(\mathscr{E}, L)N_{sub}, \qquad (2.4)$$

where m—maximum value of s.

To determine $m(\mathscr{E}, L)$ it is necessary to choose some model of the potential in the film. The simplest model, frequently used by various authors, consists of the approximation of the film by a square potential well with infinitely high walls. Such a model cannot give quantitative agreement with experiment for small values of s, but it does take into account the most characteristic feature of the electronic states in the film, namely the limitation of the transverse motion of the electrons. In the square-well model with infinite walls, the single-particle spectrum has, as is well known, the form

$$\mathscr{E}_s = \frac{\hbar^2 \pi^2}{2m^* L^2} s^2. \qquad (2.5)$$

The model of an infinitely deep well, however, does not take into account the concrete band structure of the film, and also leads to a non-self-consistent solution. On the one hand, it is assumed in the model that no forces whatever act on the electron inside the film, and on the other hand the solution leads to an inhomogeneous distribution of the electric charge, i.e., to a strong Coulomb repulsion, not initially taken into account. In spite of this, this model can be used for rough estimates. This estimate, based on the well model, gives for the lower level the same result as the uncertainty principle.

The square-well model presupposes that no forces act on the electron inside the film. However, when we consider the electron motion, it is necessary to take into account the effect of other charges on the electron, whose total charge is +e. The action of this charge on the electron in question will be significant when $\lambda \gtrsim L$. The lowering of the potential energy due to the attraction of the electron to the positive charge should cause the rising of the lowest discrete level with decreasing thickness to be smaller than would follow from the square-well model.

Formula (2.4) determines the total density of states as a function of the thickness and of the energy. This dependence is governed by the fact that the number of populated subbands increases stepwise with increasing thickness, and also with increasing energy. A plot of $N_f(E)$ at a fixed thickness is shown in Fig. 2. A characteristic feature of the state density is that it differs from 0 only for energies larger than $\mathscr{E}_1$[31,7] (the energy is reckoned from the bottom of the electron band in the bulky sample). The existence of a minimum energy is connected with the uncertainty principle.
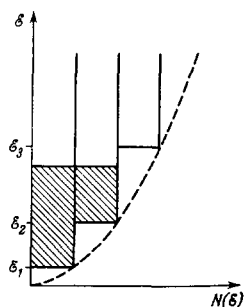
FIG 2

The distance between the neighboring steps $\mathscr{E}_{s+1} - \mathscr{E}_s$ depends, naturally, on the quantization law. If we assume that

$$\mathscr{E}_s = \frac{\hbar^2 \pi^2 s^2}{2m^* L^2},$$

then the density of states in the film at $\mathscr{E} = \mathscr{E}_s$ equals the density of states in the bulky sample at the same energy.

It is of interest to consider the behavior of the density of states when the film thickness is varied. It is this dependence which determines, in the final analysis, the experimentally observed oscillatory character of the dependence of the kinetic coefficients of thin films on their thickness. With increasing thickness, the density of states N_f at the Fermi level decreases as $1/L$ (see (2.3) and (2.4)), so long as the number of filled subbands does not change. However, at certain values of the thickness L_s, when a new subband begins to be filled, the density of the states increases jumpwise by an amount $N_{sub}(L_s)$. This results in an oscillatory dependence of the density of states on the film thickness (Fig. 3).

The decrease of the density of states, proportional to $1/L$, is not connected with the model of the film potential. The concrete form of this potential affects only the positions of the jumps on the $N_f(L)$ curve.
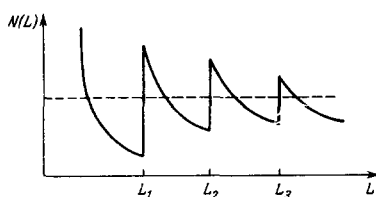

FIG. 3

3. Carrier Density and Fermi Level

Let us consider first a semiconducting film with one type of carrier[7]. The electron density in the conduction band, averaged over the film thickness is given by

$$n(T) = \frac{1}{V} \sum_s \int_{\mathscr{E}_s}^{\infty} \frac{G_{sub}(\mathscr{E})\, d\mathscr{E}}{\exp\left\{\frac{\mathscr{E} - \mu_F}{kT}\right\} + 1}, \qquad (2.6)$$

where G_{sub} is the density of states in the subband (2.2). The integration is over the s-th layer, and the summation is over all the layers.

To find the Fermi energy, we use the equation of

neutrality of the semiconductor. If the conditions for the filling of one subband are satisfied, then, assuming that the impurity levels are weakly ionized, we obtain an expression for the chemical potential

$$\mu_F(T) = -\frac{E_d}{2} + \frac{kT}{2} \ln NL \cdot \frac{\pi \hbar^2}{m^*} \cdot \frac{1}{kT}. \qquad (2.7)$$

here N—number of impurity atoms per cm^3 and E_d—activation energy of the donors, reckoned from the bottom of the band in the film. This expression differs from the corresponding formula for bulky semiconductors in the stronger dependence of μ_F on T. It is necessary to substitute in this formula[27] the activation energy determined for a given film thickness. The $E_d(L)$ dependence is due both to the rising of the bottom of the conduction band with decreasing L,[31] and to the change of the position of the impurity level which is significant if $L \lesssim a_B$ (a_B—radius of the Bohr orbit of the impurity electron).

In the approximation under consideration, we obtain for the electron concentration in the conduction band the formula

$$n(T) = \frac{1}{L}\left(\frac{m}{\pi \hbar^2}\right)^{1/2}(kT)^{1/2} \exp\left\{-\frac{E_d}{2kT}\right\}. \qquad (2.8)$$

The statistics of the carriers in semimetallic films was considered by Lutskiĭ and by Sandomirskiĭ[22]. In this case, the lowering of the edge of the valence band and the rise of the bottom of the electron band with decreasing thickness can lead to the formation of an energy gap, i.e., to a semimetal-dielectric transition at a certain thickness L_1. In the approximation of an infinitely deep well, this transition should take place at a thickness

$$L_1 = \frac{\pi \hbar}{\sqrt{2\Delta}}\left(\frac{m_e + m_h}{m_e m_h}\right)^{1/2},$$

where Δ—overlap of the energy bands in the macroscopic sample, and m_e and m_h—masses of the electron and hole in the transverse direction. For a Bi film this would yield $L_1 \sim 300$ Å. However, as already noted, the rise of the lower level in the conduction band (or lowering in the valence band) with decreasing film thickness should be much smaller in the actual film. Therefore the transition into the dielectric state could be observed only at thicknesses much smaller than the thicknesses obtained in the infinitely deep well model. Actually, it follows from experiment (see below) that in Bi films, up to thicknesses $L \sim 300$ Å, the transition into the dielectric state is not observed. Nor was this transition observed for thinner films ($L \sim 200$ Å). However, the structure of the films of such thickness is imperfect and the conclusions become unreliable.

The position of the level in the semimetal is determined from the condition of equality of the numbers of electrons and holes. In the case of an isotropic quadratic dispersion law (in the bulky sample) for electrons and holes, in the presence of strong degeneracy, the position of the Fermi level in the film does not depend on the thickness[22]. This is connected with the fact that the distance between the energy levels is inversely proportional to the mass, and the density of states is directly proportional to the mass; consequently the number of filled electron and hole subbands is always the same.

In the actual case, on the other hand, when the dispersion law is anisotropic, the numbers of the filled electron and hole subbands are different and, as follows from the neutrality conditions, proportional to the factor $\left(r \dfrac{m_{\parallel}}{m_{\perp}}\right)^{1/3}$, where r—number of equivalent valleys in the bulky sample. In particular, for Bi films the number of populated hole subbands is approximately six times larger than that of the electron subbands. The Fermi level, with allowance for the anisotropy, will oscillate weakly with change of the film thickness.

The carrier density in the semimetals must also depend on the film thickness. As shown by Sandomirskiĭ[22], with decreasing thickness the carrier density oscillates weakly even in the isotropic case (the magnitude of the relative oscillation is of the order of 1%).

4. Possibility of Experimentally Determining the Discrete Energy Levels $\mathscr{E}_S$ in Experiments on Tunneling

One of the possible methods of direct experimental observation of the quantization of the transverse-motion energy in a film is the use of tunneling of the electrons in a sandwich made of the investigated films[10].

The energy level scheme of the setup for the study of the quantization using tunneling is shown in Fig. 4. Films with a quasidiscrete energy spectrum are separated by a potential barrier, which can be realized by means of a dielectric, particularly an oxide layer.

When the external field is turned on, the energy levels of one film shift relative to the levels of the other film. If it is assumed that in the tunnel transitions the longitudinal momentum is strictly conserved, the current through the barrier will arise whenever the populated levels of one film lie opposite the free levels of the other film. As a result, a current-voltage characteristic in the form of a series of peaks is obtained. Transitions with change of the quasimomentum (phonon transitions) lead to the existence of a current even at voltages which do not satisfy the condition $eV = \mathscr{E}_{S+1} - \mathscr{E}_S$. However, as is well known, the probability of phonon transitions is much lower than the probability of the direct transitions, i.e., transitions without change of the quasimomentum. Therefore the form of the current-voltage characteristic remains in principle the same as before, but the peaks become smeared out.

Singularities on the current-voltage characteristic were observed in[16] at voltages equal to the energy difference between the discrete levels. It is highly improbable that these singularities are connected with discrete levels of the gas molecules in the gap, since the
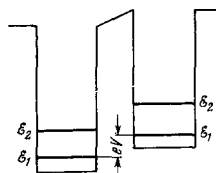
arrangement of the singularities on the current-voltage characteristic agree well with the values of the discrete levels determined in the preceding experiments[12,13]. In[16], however, the peaks on the current-voltage characteristic were not observed, while only the derivative dI/dV oscillated at values of V satisfying the condition $eV = \mathscr{E}_{S+1} - \mathscr{E}_S$. The absence of peaks on the observed current-voltage characteristic is apparently connected with the fact that under the experimental conditions the quantization took place only in one film of the sandwich.

III. INFLUENCE OF QUANTIZATION OF THE TRANSVERSE CARRIER MOTION ON THE ELECTRIC CONDUCTIVITY OF THIN FILMS

The change of the state density, and also of the form of the electronic wave functions in the transition to a thin film should affect the kinetic characteristics. Another important factor is that specific scattering mechanisms, connected with surface defects, the special character of the oscillations of the crystal lattice, etc., appear in the film. Therefore, the character of the scattering in the film changes, and leads to transitions between the stationary film states both within the subbands and between the subbands.

1. Relaxation Time and Mobility of Carriers in the Film

The most interesting feature of the electric properties of thin films is the oscillatory dependence of the mobility on the thickness[8,12,13]. This dependence is connected with the fact that mobility μ in the degenerate case is determined by the density of the states on the Fermi surface, which, as already noted, oscillates with varying film thickness. The change of the matrix elements of the transitions with decreasing film thickness, generally speaking also influences the form of the function $\mu(L)$, but the oscillatory character of this dependence is not disturbed. Indeed, the relaxation time of an electron with a planar momentum k, situated in the s-th subband, can be represented in the form of the sum

$$\frac{1}{\tau_s} = \sum_{s'} \frac{1}{\tau_{ss'}}, \qquad (3.1)$$

where τ_{ss}' is the relaxation time connected with the transitions to the s'-th subband, and the maximum value of s' is equal to the number of populated subbands. Starting from the two-dimensional kinetic equation, it can be shown that in elastic scattering the relaxation time has the same form as in the three-dimensional case[27,28].

In the case of degeneracy, with decreasing thickness, the number of terms in (3.1) increases by 1 for each new subband which begins to be populated. This leads to a jump in the relaxation-time and of the mobility, since $|M_{ss'}|^2$ is a smooth function of L. In the case when the matrix element does not depend on k and on the number of the subband (see below), the thickness dependence of $1/\tau_{ss}'$ is determined by the factor $1/L$ which arises in (3.1) on going over from summation to integration. The subsequent summation in (3.1) leads to the oscillatory dependence of $1/\tau_s$ on the film thickness.

Let us now proceed to consider concrete scattering mechanisms. An analysis of the electron-phonon interaction in a semiconducting film, for the case when one subband is populated (see conditions (1.2) and (1.3)) is

FIG. 4

presented in[27,29]. The character of this interaction depends not only on the specific feature of the electronic states, but also on the form of the phonons in the film. Generally speaking, the phonon spectrum in the film is quantized in the same way as the electron spectrum. The distance between the individual phonon branches $\omega_s(q)$ (q—two dimensional vector) is of the order of

$$\frac{\pi c}{L} \sim \frac{\theta}{nh},$$ (3.2)

where c—speed of sound, θ—Debye temperature, n—number of atomic layers in the film. However, if the film lies on a substrate, the character of the oscillations depends on the ratio of the elastic constants of the film and the substrate.

If the elastic constants of the film and of the substrates are nearly equal, the phonons in the film can be regarded as the same as in the bulky sample. A case may also occur in which the film is completely free or is very weakly connected to the substrate. In such a case the phonons are determined from the solution of the equation of the oscillations of a plate with free boundaries.

If the phonons are assumed to be the same as in the bulky sample, then we obtain for the matrix element the following expression:

$$|M_{kk'}|^2 = \frac{E^2 k_B T}{2\rho V c_l^2} \chi(q_z),$$ (3.3)

where

$$\chi(q_z) = \frac{\pi^4 \sin^2 q_z l}{q_z^2(q_z)},$$

c_l—velocity of longitudinal waves, ρ—density of the film material and E—constant of the deformation potential. The presence of the factor $\chi(q_z)$ in (3.3) is connected with the nonconservation of the transverse quasimomentum, due to the limited dimensions of the film. Since one subband is populated, the relaxation time is determined by formula (3.1) in which s = s' = 1. When the summation over q_x and q_y is replaced by integration, a factor 1/L arises, and therefore $1/\tau \sim 1/L$. To estimate the coefficient of $1/\tau$ the summation over q_z will also be replaced by integration (if the surface layer is considered, then there is no quantization of q_z, and such an approach is rigorously correct). We obtain finally

$$\frac{1}{\tau} = \frac{3}{L} \frac{m^* E^2 k_B T}{h^3 \rho c_l^2}.$$ (3.4)

As seen from (3.4), the character of the dependence of the number of collisions on the film thickness is the same as for the state density. It is also important that τ does not depend in the film on the quasimomentum, unlike the bulky sample, where $1/\tau \sim |k|$ in the scattering by phonons. This is perfectly natural, since the ratio τ/τ_{pass} should be a function of the dimensionless quantity Lk. We note, to avoid misunderstandings, that formula (3.4) is valid when $L \gg a$ (a—lattice parameter) and, on the other hand, L is bounded from above by the conditions for the filling of one layer.

Inasmuch as τ is the same for all the electrons, the mobility μ is proportional to τ, i.e., $\mu \sim L$. Thus, even in an "ideal" film, i.e., without allowance for scattering by surface defects, the mobility decreases with decreasing thickness.

The electron relaxation time in the film, upon popu-

lation of two subbands, was determined by Iogansen in[59].

In the case of a thin film with free boundaries, it is necessary to take into account the specific features of the phonon states. These features are most clearly pronounced at low temperature

$$T < \frac{\theta}{n}$$ (3.5)

(θ—Debye temperature, n—number of atomic layers in the film), when only the lower branches of $\omega_s(q)$ are excited: flexure waves, and longitudinal and transverse waves. The largest contribution to the resistance is made by longitudinal waves. As shown in[29], in this case also the relaxation time τ is proportional to the film thickness.

At higher temperatures, when condition (3.5) is not satisfied, it is necessary, generally speaking, to also take into account the higher phonon branches. The dispersion relations and the densities of the states for all the possible types of waves in films were obtained in[30].

If several subbands are populated in the case of degeneracy, the mobility oscillates with changing thickness in approximately the same manner as the density of states.

The scattering of the electrons in films with a quasi-discrete spectrum by point defects was investigated by Sandomirskiĭ[8,22]. It is shown in[8] that the relaxation time due to the elastic scattering by randomly distributed centers with δ-like potential given by

$$V(r) = \sum_{j=1}^{N} u\delta(r - R_j)$$

(R_j—coordinate of the scattering center, N—number of scattering centers in the volume of the film) is proportional, in the case of population of one subband, to the film thickness L.

Sandomirskiĭ also found the dependence of the relaxation time and of the electric conductivity on the thickness of the semimetallic film for an arbitrary number of filled electronic and hole subbands; it was likewise assumed that the scattering is by a δ-like potential. In such a model, the relaxation time is of the form[22]

$$\frac{1}{\tau_{sh}} = \frac{Nu^2}{2} m(L),$$

where m(L)—number of filled subbands.

Thus, with changing thickness, when a new subband begins to be populated, the number of possible transitions increases jumpwise, leading to the oscillatory dependence of τ on L. Since in this case τ is the same for all the electrons on the Fermi surface, τ is proportional to μ and the dependence is the same as $\tau(L)$.

The electric conductivity of thin semiconducting films was considered also by Bezak[32], who took into account only the dependence of the Fermi energy on the thickness. However, the dependence of the mobility on thickness, which is the main contribution to the conductivity oscillations, was not taken into account in[32].

The resistivity of the film oscillates not only with varying thickness, but can also oscillate with varying longitudinal electric field. The resistance oscillations can appear at such values of the field, when the energy acquired by the electron with a mean free path length is

equal to the distance between the quantum levels. This effect was considered by Kulik[33] under the assumption that quantum size effects introduce a small correction to the resistance. It is evident that the analysis made in[33] is applicable when many subbands are populated (metallic and relatively thick semiconducting films).

2. Comparison with Experiment

As already noted, in the experiments made on bismuth films[15], an oscillatory dependence of the resistance and of the mobility on the thickness was observed (Fig. 5). Inasmuch as the effective mass of the holes in the transverse direction in the film is practically two orders of magnitude larger than for electrons, the distances between the hole subbands are considerably smaller than between the electron subbands. Consequently, the quantization conditions (1.2) and (1.3) for holes are more stringent. In addition, the number of populated hole subbands, even if they are not smeared out, as already noted, are always 6—7 times larger than the number of filled electron subbands. Therefore the kinetic characteristics are essentially influenced only by the quantization of the electron motion. The Hall mobility μ at $n_p = n_e$ is equal to $\mu_h - \mu_e$. With the increase in L at thicknesses corresponding to the start of the filling of a new subband, when a jump of the density of states of the electrons takes place (see Fig. 4), μ_e decreases jumpwise, which leads to an increase in the Hall mobility (a positive sign of the Hall constant signifies that the hole mobility predominates in the film). If there were no smearing of the levels in the subbands, then on the $\mu(L)$ curve, at a thickness corresponding to the start of the filling of the new subband, there should occur an abrupt jump by a finite amount, as in the case of the state-density curve. However, no abrupt jumps are observed on the experimental curves, this being connected with the smearing of the levels by an amount on the order of h/τ. At relatively large thicknesses, when several electron subbands are populated, the square-well model is relatively satisfactory and the chemical potential depends little on the thickness. This causes the population of the new subbands to occur at equal intervals of L. As seen from Fig. 5, this is precisely the arrangement of the maxima observed experimentally. An exception is the first interval, this being apparently connected with the deviation from the chosen model at small film thickness.

At small thicknesses, a decrease of μ is observed on the plot with decreasing thickness, in agreement with the theory, which predicts a linear dependence $\tau \sim L$ for different scattering mechanisms, if one subband is filled. As was to be expected, the maxima of μ correspond to minima of the resistance on the plots of Fig. 5. The plot of $\rho(L)$ is determined not only by the mobility but also by the dependence of the concentration on the thickness, which we shall consider in greater detail in Chapter 4 in connection with the Hall effect. All the oscillations are more strongly pronounced at lower temperatures, this being connected with the better realization of the quantization conditions.

The temperature dependence of the resistivity and the Hall constant of Bi films with different thicknesses were investigated in[14]. For the entire temperature
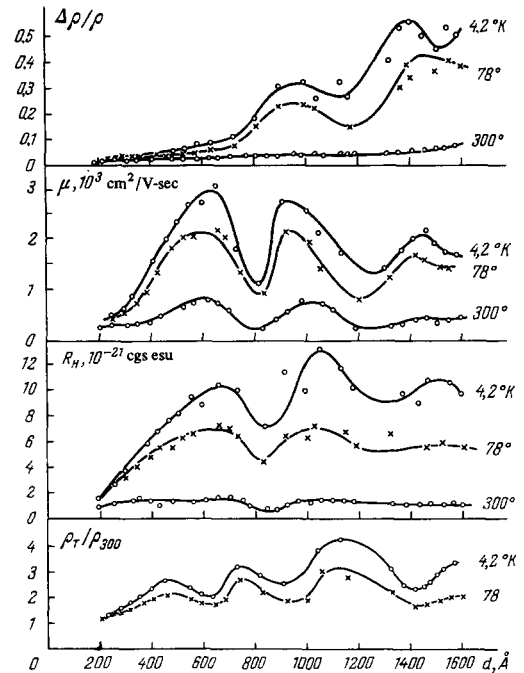

FIG. 5

interval (T ~ 4.2—200°K), the resistivity was an oscillating function of the thickness. It is important to note here that the period of the oscillations was independent of the temperature. This period coincides with the period obtained in[12,13]. It was confirmed in[14] that the large magnitude of the oscillations is determined by the dependence of the electron mobility on the thickness.

An unusual dependence of the electric conductivity and of the magnetoresistance in Bi films on the thickness was observed in[61], and was ascribed by the authors to size quantization. However, this explanation is not unambiguous, since the oscillations were not experimentally observed. This is apparently connected with the fact that the experiments were made at room temperature. The quality of the films was not investigated in[61].

As already noted in Chapter I, quantization of the resistance was observed in antimony film[17]. In view of the fact that in Sb the effective masses are relatively large, the period of the oscillations should be much smaller than in bismuth. It follows from the experiment that the period of the oscillations is approximately constant in the entire investigated interval of thicknesses and is equal to ~30 Å. No measurements of the Hall constant were made in[17], and therefore criterion (1.1) cannot be verified. The criterion (1.3) was satisfied experimentally, owing to the small investigated thicknesses, at temperatures lower than 600°K, and oscillations took place even at room temperatures.

The authors of[17] attempt to determine, by starting from the obtained period of the oscillations, the relationship of the electron and hole masses resulting in quantization. However, the connection between the period of the oscillations and the masses which they use is correct in the isotropic case[22], i.e., when an equal number of hole and electron subbands is populated at

any thickness. In fact, however, the dispersion relations $\mathcal{E}(k)$ are quite complicated, and anisotropy cannot be neglected. A correct allowance for anisotropy of the effective masses and for the different numbers of the electron and hole valleys in antimony makes the number of the populated electron and hole subbands different. For the values of mass, given in[56], the estimate by means of $(r\, m_{\parallel}/m_{\perp})^{1/3}$ (see Chapters II and III) shows that the number of filled electron subbands is approximately twice that of the hole subbands. The fact that a single period of the oscillations is clearly manifested in the experiments explicitly indicates that these oscillations are connected with the quantization solely of one type of carrier, apparently electrons. This may be connected with the fact that the quantization condition (1.1) is not satisfied for holes, or else that holes make a small contribution to the conductivity.

IV. THIN SEMIMETALLIC AND SEMICONDUCTING FILMS IN A MAGNETIC FIELD

If a thin film is placed in a magnetic field, then the quantization of electron motion will be due both to the limited size of the sample and to the presence of Landau levels. In a transverse field, the transverse motion is independently quantized as a result of the limited size of the film, and the longitudinal motion is quantized as a result of the presence of the magnetic field. Hence the spectrum becomes purely discrete, and the energy levels are produced as a result of superposition of film levels and Landau levels. On the other hand, in the case of a longitudinal field it is impossible to separate the influence of the limited transverse dimensions of the film and of the magnetic field. In consequence of the joint action of these factors, the transverse motion of electrons becomes quantized, and the longitudinal motion remains quasiclassical as before, but an anisotropy of the effective masses arises in this case.

Thus, when a magnetic field is superimposed on a film, the spectrum becomes discrete or quasidiscrete. This circumstance also must lead to oscillations of the kinetic and thermodynamic quantities. In the present case, however, the oscillations are possible both when the film thickness changes and when the field changes. If the film is sufficiently thick, then as the field varies the oscillations are transformed into the well-known oscillations of the de Haas van Alphen type for the diamagnetic susceptibility, of the Shubnikov—de Haas type for the conductivity in the magnetic field, etc.

1. Oscillations of the Magnetic Susceptibility in a Thin Film

The diamagnetic properties in a longitudinal field were first considered by Kosevich and Lifshitz[2], and the quantization was investigated both quasiclassically and by exact solution of Schrödinger's equation with a specially chosen film potential. For semimetals and semiconductors, when the lower discrete levels are filled and the quasiclassical approximation does not hold, the second case is of greatest interest. If the potential in the film is approximated by a parabolic potential well

$$V(r) = \frac{m\omega_0^2}{2} z^2.$$

(the parameter ω_0 increases with increasing thickness; an estimate can be obtained from the uncertainty relation $\hbar\omega_0/2 = (\pi/L)^2 \hbar^2/2m$), and this yields the following spectrum:

$$\mathcal{E} = \left(M + \frac{1}{2}\right) \hbar\omega_c + \frac{\hbar^2 k_x^2}{2m} + \frac{\omega_0^2}{\tilde{\omega}^2} \frac{\hbar^2 k_y^2}{2m}, \tag{4.1}$$

where $\tilde{\omega} = \sqrt{\omega_c^2 + \omega_0^2}$ is the field directed along x, and ω_c is the cyclotron frequency.

From (4.1) we obtain a formula for the period of the oscillations when the magnetic field is varied:

$$\Delta\left(\frac{1}{H}\right) = \frac{2\mu_B}{\hbar} \left(\frac{\tilde{\omega}}{\omega_c}\right)^2 \tag{4.2}$$

(μ_B—Bohr magneton). In the limiting case of a macroscopic crystal ($\omega_0 = 0$), we obtain the usual formula.

At a fixed film thickness, the period of the oscillation $\Delta(1/H)$ decreases with increasing magnetic field. This is connected with the fact that the distance between the "magnetofilm" levels depends less on the magnetic field than the intervals between the Landau levels in the bulky sample.

It is important to note that in a film, the criterion for the existence of the de Haas—van Alphen effect becomes less stringent. In a bulky sample, the magnetic field should be sufficiently strong to satisfy the conditions

$$\omega_c\tau \gg 1, \quad \hbar\omega_c \gg k_B T. \tag{4.3}$$

In a film, the role of the cyclotron frequency is played by the "magnetofilm" frequency $\tilde{\omega}$, and the criterion takes the form

$$\tilde{\omega} = \sqrt{\omega_0^2 + \omega_c^2} \gg \frac{1}{\tau}, \quad \hbar\tilde{\omega} \gg k_B T. \tag{4.4}$$

At relatively small thicknesses $\tilde{\omega}$ is determined essentially by the value of ω_0. Therefore, the de Haas—van Alphen criterion is satisfied even in weak magnetic fields.

The oscillation period, however, will be large at large values of ω_0, since the change of the magnetic field will have a relatively small effect on the magnitude of $\tilde{\omega}$. Indeed, in order for oscillations to take place, it is necessary that the upper filled magnetofilm level rise by $\hbar\tilde{\omega}$. This rise is equal to the product of the change of the distance between neighboring levels

$$\hbar\omega_c \frac{\Delta\omega_c}{\omega_0}$$

and the number of filled levels. Consequently

$$\Delta\omega_c \sim \frac{\hbar\omega_0^3}{\omega_c\mu_F}.$$

For acceptable values of the period of the oscillations $\Delta(H^{-1})$ it is necessary that the number of filled levels $\mu_F/\hbar\omega_0$ be not very small. We present estimates for Bi films, for which film levels were observed. At a thickness $L \sim 2000$ Å and $H \sim 10^3$ G ($\omega_c \sim 0.3\,\omega_0$) we obtain for the oscillation period $\Delta\omega_c$ a value on the order of the cyclotron frequency ω_c itself.

Oscillations of the magnetic susceptibility occur also when the thickness of the film L changes. These oscillations, however, arise only in a definite thickness interval. At large L, the criterion for the de Haas—van Alphen effect is not satisfied if the magnetic field is insufficiently strong ($\omega_c \ll 1/\tau$). On the other hand, if the magnetic field is strong ($\omega_c > 1/\tau$), then the oscillations do not arise for another reason. The level shift

will be insufficient, since the change of the thickness has little effect on $\tilde{\omega}$. With decreasing thickness, the effect disappears when only one subband is populated. This minimum value of the thickness increases with increasing magnetic field.

For the intermediate region of thicknesses, the period of the oscillations can be readily obtained from qualitative considerations described above.

If we assume in addition that $\omega_C \ll \omega_0$, then we obtain

$$\frac{\Delta L}{L} \sim \frac{\hbar\omega_0}{\mu_F}. \tag{4.5}$$

For the presented parameters in the Bi film we have $\Delta L \sim 200$ Å.

We now proceed to consider diamagnetism for the case when the field is perpendicular to the film. At this field orientation, the energy spectrum of the electrons is a superposition of film and magnetic energy levels $\mathscr{E} = \mathscr{E}_s + (M + 1/2)\hbar\omega_C$. At a constant thickness, the change of the magnetic field leads to a non-simultaneous crossing of the Fermi surface by levels with different values of s. Therefore, in a film, for an arbitrary form of the film potential, there will occur a decrease of the amplitude of the oscillations, and the periodicity in their arrangement will become violated. On the other hand, if only one film level is populated, then the violation of the periodicity will not take place. The period of the oscillations is in this case

$$\frac{\Delta\omega_c}{\omega_c} \sim \frac{\hbar\omega_c}{\mu_F - \mathscr{E}_1}$$

and increases with decreasing L.

The oscillations of the diamagnetic susceptibility will occur also when L varies, since the number of the populated magnetic levels changes stepwise at certain values of the thickness.

Singularities in the state density in the film also lead to oscillations of the spin paramagnetic susceptibility[7]. The origin of this effect can be readily understood from the following considerations. Let the Fermi level in the absence of a magnetic field lie between the levels $\mathscr{E}_s$ and $\mathscr{E}_{s+1}$. Under the condition of total degeneracy $k_B T < \mathscr{E}_{s+1} - \mathscr{E}_s$, then the magnetic moment is

$$M = \int_{\eta - \mu_B H}^{\eta + \mu_B H} N_f(\mathscr{E})\, d\mathscr{E}. \tag{4.6}$$

As long as the magnetic field is weak, so that $\eta \pm \mu_B H$ lies on one step of the interval $(\mathscr{E}_s, \mathscr{E}_{s+1})$, the magnetic moment will be proportional to the field, since the state density is constant in this energy interval. If crossing of the level $\mathscr{E}_s$ takes place with increasing field, then the magnetic susceptibility decreases jumpwise, and when the level $\mathscr{E}_{s+1}$ is crossed it increases jumpwise. With further increase of the field, the magnetic susceptibility will pulsate.

2. Galvanomagnetic Phenomena in Thin Films

We proceed to consider galvanomagnetic phenomena in thin films. A study of these phenomena is of practical interest in connection with the fact that these phenomena are used in the determination of the carrier density and the singularities of the energy spectrum of the semiconductors. The relations obtained by considering galvanomagnetic phenomena in a bulky semiconductor cannot be transferred automatically to a thin film without taking into account the specific features of the film states. We shall stop to discuss separately the case of a quantizing magnetic field and the case when the Landau quantization is not significant.

a) **Quantizing magnetic field.** In the case of a quantizing magnetic field, a realignment of the spectrum takes place as a result of the limited dimensions of the sample and of the appearance of Landau levels.

Transverse galvanomagnetic phenomena in a bulky sample were first considered by Titeica[34] and later a strict quantum theory based on the solution of the equation for the density matrix was proposed by Adams and Holstein[35]. Erukhimov and one of the authors solved this problem as applied to a film[36].

For an electron in a transverse magnetic field and longitudinal electric field, the wave function is proportional to $\exp(-(x - x_k^0)^2/l^2)$, and the spectrum is given by

$$\mathscr{E}_{Mh} = \left(M + \frac{1}{2}\right)\hbar\omega_c + \mathscr{E}_s - e\mathscr{E}x_k^0, \tag{4.7}$$

where k is in this case the projection of the quasimomentum on the y axis, the oscillation center is

$$x_k^0 = -l_k^2 + \frac{e\mathscr{E}}{m^*\omega_c},$$

the magnetic length $l = (\hbar/m^*\omega_c)^{1/2}$, and $\mathscr{E}_s$ are the film levels. The random character of the scattering, just as in a bulky sample, causes the perturbed state to be impure, and transitions arise between the stationary levels. In this case the electron system should be described by a density matrix. The Hall current j_y has a definite value already in the stationary state. The Hall constant will also equal $R = 1/ecn$, but the concentration in this case is a function of L and H. Since the dependence of ψ on x is described by a real function, the current j_x along the electric field can appear only as a result of scattering. Scattering gives rise to transitions with a change of discrete quantum numbers s and M and of the oscillation center x_k^0, which runs through a continuous set of values. Inasmuch as the wave function is localized in the interval l, the matrix elements of the scattering potential are significantly different from 0 only for those transitions for which $x_k^0 - x_{k'}^0 \sim l$. For such transitions, the electric energy should change by an amount $e\mathscr{E}l$. If the electric field is weak and the intervals between the magnetic levels and the film levels are sufficiently large, so that $\hbar\omega_c \gg e\mathscr{E}l$ and $\Delta\mathscr{E}_s \gg e\mathscr{E}l$, then the energy conservation law forbids any transitions between the stationary states. This situation is typical of a film. In a bulky sample, where the energy of the motion of the electrons along the magnetic field is continuous, transitions between the stationary states are possible at an arbitrarily weak electric field E. This circumstance makes it possible to seek an addition to the density matrix in the bulky crystal in the form of a series of powers of E, leading to Ohm's law in the case of weak fields. On the other hand, in the case of a film the current is practically nonexistent up to fields $E \lesssim E_0 = \hbar\omega_c/el$, i.e., fields in which the electron acquires an energy $\hbar\omega_c$ over the magnetic length.

Under the condition that one magnetic and one film level are filled, the calculations[36] lead to the following dependence of the dissipative current j_x on the inten-

sity E

$$j_x \sim \exp\left\{-\frac{1}{2}\left(\frac{E_0}{E}\right)^2\right\}. \tag{4.8}$$

When several magnetic levels are populated, the form of $j_x(E)$ remains qualitatively unchanged, but the parameter E_0 becomes a nonmonotonic function of H, leading to an oscillatory dependence of the dissipative current on the magnetic field.

For very large values of the magnetic field, when only one magnetic level is populated, this dependence becomes monotonic. In order for the magnetic field to be quantizing, it is necessary to satisfy the conditions

$$\frac{\hbar}{\tau} \ll \hbar\omega_c, \quad k_B T \ll \hbar\omega_c. \tag{4.9}$$

The second of these conditions does not impose any rigid limitations on the temperature, especially when the cyclotron mass $m_c = \sqrt{m_1^* m_2^*}$ is small, as is the case, for example, in Bi films, in which quantization of the transverse motion was observed, or in InSb. The condition (4.9) is more stringent with respect to the relaxation time or its associated mobility, which in films is lower than in the bulky sample. Comparatively more promising in the sense of satisfying the conditions (4.9) are InSb films, in which, owing to the isotropy of the dispersion law, the cyclotron mass is small for any film orientation: $m_c^* \sim 0.01\, m_0$. For an isotropic dispersion law, conditions (4.9) are equivalent to the condition

$$\mu > \frac{10^8}{H}\ \text{cm}^2/\text{V-sec}.$$

In a longitudinal quantizing magnetic field[60] the situation changes radically, since the motion along the film can be regarded as quasi-classical. In considering the de Haas—van Alphen effect it was noted that the presence of a longitudinal magnetic field leads (see (4.1)) to an increase in the distance between the discrete film levels, which is effectively equivalent to a decrease of the thickness. The influence of the magnetic field on the motion along the film results, as it were, in an increase of the effective mass in a direction perpendicular to the field. When the magnetic field changes, the density of states on the Fermi surface will oscillate. In the de Haas—van Alphen effect, this has led to pulsations of the magnetic susceptibility, and in this case pulsations of the current will take place (the Shubnikov—de Haas effect). Oscillations will arise also when the thickness changes. The criterion for the existence of the effect will evidently be the same as in the case of the de Haas— van Alphen effect (4.3).

b) **Nonquantizing transverse magnetic field.** Let us consider the case when the Landau quantization can be neglected (conditions (4.9) are not satisfied). In a transverse magnetic field, if a relaxation time can be introduced, the dependence of the Hall constant R_H and of the magneto-resistance $\Delta\rho/\rho$ on τ will be the same in the film as in the bulky sample, since the dimensionality of the space is not essential in the derivation of these relations. The Hall constant and the magneto-resistance in a bulky crystal are obtained by the kinetic-equation method. On going over to a film, it is natural to assume that the kinetic equation is valid only when considering longitudinal carrier motion, which can be regarded as quasiclassical. A non-quantizing magnetic field, directed perpendicular to the film, does not violate the quasi-

classical character of the longitudinal motion. Hence the kinetic-equation method can be used to calculate the Hall effect and the magnetoresistance along the film in a transverse magnetic field.

If the magnetic field is weak ($\omega_c \ll 1/\tau$), then R_H and $\Delta\rho/\rho$ are determined, as is well-known, by the relations

$$R_H = -\frac{\overline{\tau^2}}{\overline{\tau}^2}\frac{1}{enc}, \quad \frac{\Delta\rho}{\rho} = \left(\frac{eH}{m^*c}\right)^2\frac{\overline{\tau^3}\overline{\tau}-\overline{\tau^2}}{\overline{\tau^2}} \tag{4.10}$$

(the averaging over the electronic states is denoted by a bar).

Since it can be approximately assumed for different scattering mechanisms when one subband is filled, that τ is proportional to L, it follows from (4.10) that the Hall constant is connected with the concentration in the same manner as in the bulky sample. The magneto-resistance then decreases as L^2 with decreasing thickness[22,28].

If many subbands are populated, then, in connection with the oscillations of $\tau(L)$, the quantity $\Delta\rho/\rho$ should also oscillate, and the dependence of $R_H(L)$ will be determined by the dependence of the concentration on the thickness[22].

In the case of a strong non-quantizing field ($\omega_c \gg 1/\tau$, $\hbar\omega_c \ll k_B T$), the Hall constant and the magnetoresistance are determined by the formulas

$$R_H = \frac{1}{enc}, \quad \frac{\Delta\rho}{\rho} = \overline{\tau}\left(\frac{1}{\tau}\right) - 1. \tag{4.11}$$

In this case the Hall constant again depends on the thickness only via the concentration, and the magneto-resistance does not depend significantly on L[28].

3. COMPARISON WITH EXPERIMENT

A dependence of R_H and of $\Delta\rho/\rho$ on the thickness was observed experimentally[12,13] for magnetic fields $H \sim 10^4$ G. For the experimentally-observed values of mobility ($\mu \sim 10^3$ cm^2/V-sec), these fields are weak. For magnetoresistance, the dependence on L is connected with a change of the relaxation time, just as is the case for mobility. For small thicknesses, as follows from the foregoing reasoning, $\Delta\rho/\rho$ tends to 0. For thicknesses larger than L_1 it can be assumed that degeneracy takes place, and R_H is determined under the condition $n_h = n_e$ by the formula

$$R_H = \frac{1}{enc}\frac{\mu_h - \mu_e}{\mu_h + \mu_e}. \tag{4.12}$$

As shown in[22], the concentration is an almost monotonic function of the thickness for $L > L_1$.

As was shown above (Chapters III and II), the anisotropy of the effective masses in Bi makes it possible to neglect the oscillations of μ_h in comparison with the oscillations of μ_e. Therefore, the factor $(\mu_h - \mu_e)(\mu_h + \mu_e)$ in (4.12) will oscillate with changing L, thus apparently explaining the experimentally observed oscillations of R. In real films it may turn out that the electron and hole densities are not equal. Then formula (4.12) is somewhat modified. However, if we disregard the effective-mass anisotropy, we cannot explain the strong oscillations of the Hall constant observed in[12,13].

When the film is so thin that one subband is populated, the Hall constant decreases with decreasing L, as can be seen from the experimental curve (see Fig. 5).

If we use the square-well model to approximate the quantization law, then at small thicknesses on the order of 300−400 Å[24] the overlap of the energy bands in the Bi films should vanish. This should lead to an exponential decrease of the concentration with decreasing thickness. The decrease of the concentration should cause a sharp increase of R, which is not observed experimentally. This shows that in the investigated samples no transition to the dielectric state takes place, i.e., evidently, at small thicknesses the square-well model gives a poor approximation.

V. TEMPERATURE OF TRANSITION INTO THE SUPERCONDUCTING STATE IN THIN FILMS WITH QUASIDISCRETE ENERGY SPECTRUM

The conditions for the appearance of superconductivity in films with a quantized spectrum differ from the corresponding conditions in the bulky sample. This is connected with the dependence of the electron density of states on the thickness of the film and with the special character of the Cooper pairing. The electrons forming Cooper pairs in the film should have opposite planar momenta k, just as in the bulky sample. As to the discrete quantum number s, it should be the same for both electrons of the pair, since the electrons from different subbands with a given energy have different k. Thus, the pairing of the electrons in different subbands can be neglected, at least so long as $\Delta \mathscr{E}_s \gg k_B T_c$ (corresponding to thicknesses $L \lesssim 10^{-5}$ cm).

The described picture of the Cooper pairing of the electrons in a film is analogous to the two-band model of a bulky superconductor[37], in which pairing of the electrons belonging to different subbands is neglected.

1. Case of Population of One Subband

Of the greatest interest is the case of population of only one subband (semimetals, semiconductors), when the specific character of the film state is most strongly pronounced. We note that superconductivity was recently observed in degenerate semiconductors (GeTe, SrTiO$_3$, PbTe)[27].

When electrons populate one subband, the problem in k-space becomes two-dimensional, although the film is essentially a three-dimensional formation ($L \gg a$). The usual method of finding the energy gap leads to an equation similar to the Cooper equation for the bulky superconductor:

$$1 = \frac{g}{2V} \sum_{k'} \frac{\operatorname{th} \frac{\sqrt{\Delta^2 + \xi_{k'}^2}}{2k_B T}}{\sqrt{\Delta^2 + \xi_{k'}^2}}, \qquad (5.1)$$

Here V—volume of the film. The summation in (5.1) is carried out over the two-dimensional Brillouin zone. The interaction constant g, generally speaking, differs somewhat from g in the bulky sample. This difference, however, is not fundamental, and it at least does not contain the factors 1/L, as can be seen from the expression for the interaction constant obtained on the basis of a consideration of the Frohlich Hamiltonian $g = |M_{kk'}|^2/\hbar \omega$, where the matrix element is

$$M_{kk'} = \frac{E}{V} \int \psi_{k'}^* \operatorname{div} \overline{u} \, \psi_k \, dV,$$

and ψ and u are normalized to the volume of the film.

The two-dimensional Cooper equation leads[38] to the usual relation between the critical temperature and the energy gap at absolute zero: $2\Delta = 3.52 \, k_B T$, with

$$\Delta = 2\hbar \omega_c \exp\left(-\frac{2\pi}{g} \frac{L\hbar^2}{m^*}\right). \qquad (5.2)$$

A distinguishing feature of this case is the fact that the energy gap and T_c are exponentially dependent on the film thickness. This circumstance is due to the dependence of the density of the states on L in one subband: as seen from (5.2), the argument of the exponential can be represented in the form $-1/gN(L)$. It follows from (5.2) that if a bulky semiconductor or semimetal is a superconductor, then in the film state, at sufficiently small thickness, it is possible to attain a higher value of the critical temperature. Another feature of T_c in film when only one subband is filled is the weak dependence on the carrier density. This feature is connected with the fact that the density of states at the Fermi level does not depend on the concentration, and the characteristic phonon frequency ω_c and the coupling constant g cannot give a strong dependence, since only intervalley transitions are important in superconducting semiconductors (see below). Of course, these conclusions are not valid for very small concentrations when the electron system is far from degeneracy. The characteristic phonon frequency ω_c is determined by the mean value of the momentum of the virtual phonon. Attraction between electrons corresponds to virtual-phonon momenta on the order of the difference between the momenta of the interacting electrons.

In this connection, Cooper pairing of electrons from one valley in semiconductors and semimetals will correspond to a small phonon frequency, much lower than the Debye frequency. Consequently, in semiconductors with one valley it is impossible to expect a high T_c[32]. Cohen called attention to the fact that in a multivalley semiconductor, high values of the critical temperature are possible as a result of intervalley transitions[40]. In the case of a film, a similar situation is obtained, i.e., we can expect the appearance of superconductivity only in the presence of several valleys. Consequently, in formula (5.2) the characteristic phonon frequency is of the order of the Debye frequency, since intervalley transitions correspond to a momentum transfer on the order of the reciprocal lattice constant.

We have noted above that the density of states in a film at the Fermi level, when one subband is filled, does not depend on the carrier density. In this connection, we can expect the appearance of superconductivity in semiconducting films with low carrier density. Such a semiconductor, in the immediate vicinity of 0 temperature, is nondegenerate (the impurity band is narrow and does not overlap the conduction band). With increasing temperature, degeneracy may set in. The possibility of occurrence of superconductivity of semiconductors in the presence of such a situation was investigated by Kogan and one of the authors[41]. It was shown there that superconductivity can appear in the temperature interval bounded from both above and below.

The existence of a lower temperature limit for superconductivity T_{sup} is connected with the fact that when $T < T_{sup}$ the semiconductor is far from degeneracy and no Cooper pairing takes place.

2. Case of a Metal Film

The peculiarities of superconductivity at large carrier density, when several energy subbands are filled in the film, were first considered by Blatt and Thompson[42]. They solved numerically the three dimensional Cooper equation with allowance for the quantization of the electron energy, for fixed parameters characteristic of metals. In[42] no account was taken of the special character of the Cooper pairing of the electrons in the film, so that the obtained oscillations of the critical temperature with varying thickness are due entirely to the effect of pulsation of the density of states. The influence of the dependence of the density of states on the thickness at T_c was investigated also in[43,44].

As was noted, Cooper pairing in a film should occur only between electrons belonging to the same subband. Hence, the system of two-dimensional equations should be of the form[45]

$$\Delta_s = \frac{g_{sl}}{2V} \sum_l \sum_{kl} \frac{\Delta_l \, \text{th} \frac{\mathcal{E}_l}{2k_BT}}{\mathcal{E}_l} , \qquad (5.3)$$

where Δ_s—gap in the s-th subband, g_{sl}—interaction constant corresponding to the virtual transitions between the s-th and l-th subbands, and $\mathcal{E}_e = \sqrt{\Delta^2 + \xi_e^2}$ is the energy of the electron in the normal metal, reckoned from the Fermi level, as in the model of a multi-band bulky metal.

The critical temperature will be the same for all subbands, although the energy gaps Δ_s, generally speaking, are different. From the system (5.3) we obtain an expression for the critical temperature:

$$T_c = 1,14\hbar\omega_c \exp\left\{ -\frac{2\pi L\hbar^2}{m^* \sum_l g_{ll}\lambda_l} \right\} , \qquad (5.4)$$

where $\lambda_e = \Delta_e/\Delta$ as $T \to T_c$.

The coupling constants g_{sl}, generally speaking are not identical. However, if we neglect their difference, the gaps Δ_s turn out to be equal and formula (5.4) becomes

$$T_c = 1,14\hbar\omega_c \exp\left\{ -\frac{1}{gN_f} \right\} , \qquad (5.5)$$

i.e., the distinction from a bulky sample will occur only in the density of states. In view of the oscillatory character of $N_f(L)$, the critical temperature will also oscillate with changing film thickness. Such oscillations can be observed experimentally, apparently, only in semiconducting films.

When the number of populated subbands is large (metallic film), the oscillations have a small period (on the order of the lattice parameter). It is therefore meaningful to speak only of the average $N_f(L)$ dependence. The average value of N_f, generally speaking, increases with decreasing L. However, this effect is small. According to estimates[44], even under the most favorable choice of the boundary conditions, leading to a maximum density of states, this effect cannot explain the growth of the critical temperature in films of certain metals, observed experimentally[46,47,48,57] (a review of certain experimental data on the variation of T_c in thin metallic films is contained in the note by Maksimov[49]).

An explanation of the increase in the critical temperature within the framework of a model with quasi-discrete spectrum, due to the difference in the constants,

was carried out in[50,51]. It is shown in[50] that the average value of the interaction constant, when the film thickness is reduced by $\frac{1}{2}$, increases by a factor $(1 + a/L)$, leading to a growth of the critical temperature in accordance with the formula

$$T_L/T_{2L} \approx (\hbar\omega_c/k_BT_{2L})^{a/L},$$

where a is on the order of the lattice constant. Estimates made for Al films are in good agreement with the experiment.

Another possible explanation for the increase of T_c in aluminum films was given by Strongin and Paskin[47] on the basis of the non-phonon mechanism of superconductivity, connected with the interaction of the electrons with neutral molecules on the surface, first proposed by Ginzburg[52]. From the point of view of the indicated mechanism, the increase of T_c in the film can be attributed to the interaction of the electrons with the oxide on the surface. In this connection, of interest are the experiments of Alekseevskiĭ and Mikheeva[48], in which the critical temperature in aluminum films was independent of the thickness of the oxide layer.

The quantization effects can be significant in those metals in which there exist groups of electrons and holes with small effective masses and small concentrations. In particular, such a feature is possessed by the Fermi surface in Al[53]. Recent experiments performed by Alekseevskiĭ and Vedeneev on photoabsorption[58] apparently confirm the presence of a discrete spectrum in aluminum films.

The problem of determining the critical temperature and the spectrum of single-particle excitations for the case when the interaction of the electrons is nonhomogeneous over the thickness of the film was considered in[54]. It was shown that the results coincide with the case when the interaction is homogeneous over the thickness, with an accuracy up to a replacement of the interaction constant by its mean value. The relations for T_c and Δ obtained in[54] follow from the results of[45,50] if the coupling constants for the different subbands can be regarded as identical. In particular, in this case the energy gaps pertaining to different subbands, as already noted, will be the same

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The experimental material accumulated to date proves convincingly the existence of quantum size effects in thin films. However, the range of investigated materials is still quite small. This is connected, on the one hand, with the difficulties of obtaining perfect films, and also with the fact that until recently the number of experimental investigations aimed at observing quantum size effect in semiconducting and semimetallic films was relatively small.

The experimenters are now being offered much greater possibilities as a result not only of progress in the technology of film preparation, but also as a result of recent theoretical research, which has predicted many effects accessible to observation. In addition to the already observed pulsations of the kinetic coefficients with varying film thickness, it has been shown by now that size quantization greatly influences such phenomena as the de Haas—van Alphen effect, the Shubnikov—de Haas effect, and the superconductivity of semiconducting films. Size quantization can also lead to a noticeable

increase of the width of the forbidden band in semiconductors and the conversion of a semimetal into a dielectric, to the appearance of resonant absorption of light in films, to an oscillatory dependence of the film resistance on the longitudinal electric field, etc.

To obtain more detailed information concerning the characteristics of real films it is necessary to develop effective methods of obtaining the dispersion law in the presence of quantization from the experimental data.

In connection with the progress noted recently in the procedures of obtaining perfect semiconducting and semimetallic films, we can expect the number of investigated materials to increase. There are promising searches for quantum effects in films of III-V semiconductor compounds, in which the effective mass is small and the mobility is large (for example, InSb). To observe the semimetal-dielectric conversion with increasing thickness, a convenient material is an alloy of Bi and Sb, in which the overlap of the energy bands is smaller than in pure Bi, and can change with varying Sb concentration. It is also desirable to perform experiments with films of superconducting semiconductors (for example, GeTe).

One can hope that quantum size effects will turn out to be important also in the study of near-surface regions of semiconductors.

Quantum size effects are not only of independent interest, but in principle can be useful for the determination of the parameters of bulky crystals. However, no suitable procedure has yet been developed. Very promising is the creation of film instruments, the operating principle of which is based on the use of different quantum effects[10,62-65].

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