QUANTUM SIZE EFFECTS AT HIGH TEMPERATURES

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Quantum size effects at temperatures $T \sim \zeta$ are investigated theoretically. For semimetals, the chemical potential is $\zeta \sim 100^\circ K$ and depends little on T. It is shown that the amplitudes and phases of the size-effect oscillations of the thermodynamic quantities have a unique temperature dependence that is due to the non-equidistant distribution of the quantum energy levels. The obtained temperature dependence of the oscillation amplitudes explains qualitatively why the experimentally observed[5,6] decrease of the oscillations with increasing temperature is slower than called for by the theory[1,2].

1. INTRODUCTION

It is known[1,2] that spatial quantization of the conduction-electron energy in a film gives rise to periodic oscillations of the thermodynamic and kinetic quantities as functions of the film thickness L. Experimental investigations[3-6] of quantum size effects (QSE) in bismuth and antimony films show clearly the presence of periodic oscillations of the physical quantities not only at low temperatures $\pi T/\zeta \ll 1$, but also at temperatures $\pi T/\zeta \sim 1$. For semimetals, the chemical potential ζ depends little on the temperature and its order of magnitude is $\zeta \sim 100^\circ K$.

We shall arbitrarily regard temperatures $\pi T/\zeta \gtrsim 1$ as high. In this temperature region, the formulas of the theory[1,2] of the QSE, obtained under the assumption that $\pi T/\zeta \ll 1$, may, naturally, not hold. It is therefore of interest to report the results of a theoretical investigation of the QSE at arbitrary temperatures.

We calculate in this paper the oscillating part of the thermodynamic potential Ω_{osc}. Knowing Ω_{osc}, we can find the oscillations of the other thermodynamic quantities.

We show that at high temperatures the periodic oscillations of the physical quantities are possible only for films that are not too thin. The dependence of the amplitude and of the phase of the oscillations on the temperature is connected strongly with the non-equidistant character of the distribution of the quantum energy levels; an investigation of this character may yield valuable information on the structure of the electronic energy spectrum of the films.

2. QUANTUM OSCILLATIONS OF THERMODYNAMIC QUANTITIES

According to the general formulas of statistical physics, we have for the thermodynamic potential Ω of the electrons in the film

$$\Omega = -\frac{2VT}{(2\pi\hbar)^2 L}\sum_{n=1}^{\infty}\iint d^2\mathbf{p}\ln\left(1+\exp\frac{\zeta-\varepsilon_n(\mathbf{p})}{T}\right), \qquad (1)$$

where $\epsilon_n(\mathbf{p})$ is the quantized energy spectrum of the electron, $\mathbf{p}$ the quasimomentum component along the film, V the volume of the film, and the z axis is directed normal to the film.

In the case of quadratic dispersion

$$\varepsilon_n(\mathbf{p}) = \frac{1}{2m}\left(\frac{\pi\hbar n}{L}\right)^2 + \frac{p^2}{2m}, \qquad (2)$$

the oscillating part of the thermodynamic potential Ω_{osc} can be calculated in explicit form at all temperatures. As a result we obtain from (1) and (2)

$$\Omega = \tilde{\Omega} + \Omega_{osc} = \tilde{\Omega} + \sum_{k,s=1}^{\infty}\frac{1}{k^2}\Omega_{ks},$$

$$\Omega_{ks} = \frac{VT}{\pi\lambdabar L^2}\exp\left[-k\sqrt{2}\frac{L}{\lambdabar}\left(\sqrt{1+\Theta_s^2}-1\right)^{1/2}\right]\left\{\frac{1}{\sqrt{2}}\left(\sqrt{1+\Theta_s^2}+1\right)^{1/2}\right.$$
$$\times\sin\left[k\sqrt{2}\frac{L}{\lambdabar}\left(\sqrt{1+\Theta_s^2}+1\right)^{1/2}\right]+\left[\frac{1}{\sqrt{2}}\left(\sqrt{1+\Theta_s^2}-1\right)^{1/2}\right.$$
$$\left.\left.+\frac{\lambdabar}{2kL}\right]\cos\left[k\sqrt{2}\frac{L}{\lambdabar}\left(\sqrt{1+\Theta_s^2}+1\right)^{1/2}\right]\right\}, \qquad (3)$$

where $\tilde{\Omega}$ is a smooth function, $\lambdabar = \hbar/\sqrt{2m\zeta}$, and

$$\Theta_s = \pi(2s-1)T/\zeta. \qquad (3')$$

At low temperatures, $(\pi T/\zeta)^2 \ll 1$, summing over s in (3), we obtain for Ω_{osc} the well-known[1] result[1)]:

$$\Omega_{osc} = \frac{VT}{2\pi\lambdabar L^2}\sum_{k=1}^{\infty}\frac{1}{k^2}\left[\sin\left(2k\frac{L}{\lambdabar}\right)+\frac{\lambdabar}{2kL}\cos\left(2k\frac{L}{\lambdabar}\right)\right]\operatorname{sh}^{-1}\left(k\frac{\pi T}{\zeta}\frac{L}{\lambdabar}\right). \qquad (4)$$

At high temperatures, $\pi T/\zeta \sim 1$, it follows from (3) that Ω_{osc} has a rather complicated form in the case of thin films of thickness $L \sim \Delta L_s$, being the sum of oscillations with different periods ΔL_s.

For not too thin films, when all the terms in (3) with the exception of Ω_{11} can be neglected, Ω_{osc} is a periodic oscillating function of L, whose period

$$\Delta L = \pi\lambdabar\sqrt{2}\left(1+\sqrt{1+(\pi T/\zeta)^2}\right)^{-1/2}, \qquad (5)$$

depends on the temperature. The amplitude of the oscillation is proportional to $\exp(-2\pi^2 T/\Delta_{eff})$, where the function

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[1)] In the temperature region $0 < \pi TL/\zeta\lambdabar \ll 1$, the series (4) converges slowly and does not give an idea of the form of the oscillation curve. A different expansion for Ω_{osc}, which converges rapidly at low temperatures $\pi TL/\zeta\lambdabar \ll 1$, was obtained in [1] and used to analyze the temperature dependence of the form of the size-effect oscillations of the specific heat.

$$\frac{T}{\Delta\varepsilon_{\text{eff}}}=\frac{1}{\pi^2\sqrt{2}}\frac{L}{\bar{\lambda}}\left(\sqrt{1+\left(\frac{\pi T}{\zeta}\right)^2}-1\right)^{1/2} \tag{6}$$

increases with increasing T more slowly at temperatures $\pi T/\zeta \sim 1$ than at low temperatures $(\pi T/\zeta)^2 \ll 1$ (See Fig. 1).

The temperature dependence of the amplitude of the oscillations is characterized by the value of $\Delta\epsilon_{\text{eff}}$ determined from formula (6). When $(\pi T/\zeta)^2 \ll 1$ we have $\Delta\epsilon_{\text{eff}} = 2\pi\bar{\lambda}\zeta/L$, which coincides with the distance $\Delta\epsilon$ between the quantum energy levels near the chemical potential ζ. With increasing temperature, $\Delta\epsilon_{\text{eff}}$ increases and at $(\pi T/\zeta)^2 \gg 1$ we have $\Delta\epsilon_{\text{eff}} = \Delta\epsilon\sqrt{\pi T/2\zeta}$. The increase of $\Delta\epsilon_{\text{eff}}$ with increasing temperature is due to the nonequidistant character of the energy spectrum (2) and it plays an important role in the estimate of the amplitude of the oscillations at high temperatures $\pi T/\zeta \sim 1$.

The non-equidistant character of the quantum energy levels becomes manifest also in oscillations of the thermodynamic quantities with changing temperature. The period of the temperature oscillations, as follows from (3), is

$$\Delta(T^2)=\zeta^2\frac{2^{5/2}}{\pi}\frac{\bar{\lambda}}{L}\sqrt{1+\left(\frac{\pi T}{\zeta}\right)^2}\left(1+\sqrt{1+\left(\frac{\pi T}{\zeta}\right)^2}\right)^{1/2}. \tag{7}$$

When $(\pi T/\zeta)^2 \ll 1$ we get $\Delta(T^2) = 8\bar{\lambda}\zeta^2/\pi L$ from (7). In determining the period (7) of the temperature oscillations, we have neglected the temperature dependence of the chemical potential ζ. For semimetals at $\pi T/\zeta \sim 1$, the temperature dependence of the phase of the oscillations is determined mainly by the non-equidistant character of the energy spectrum. In more detailed calculations, if the explicit $\zeta(T)$ dependence is known, the formulas (3)—(6) above enable us to take into account also effects connected with the change of the chemical potential ζ with changing temperature.

For bismuth films of thickness $L \sim 10^{-5}$ cm at temperatures $T \sim 100°$K, the period of the temperature oscillations is $\Delta T \sim 10°$K. To observe these oscillations, it is necessary to have not too thin films with $L > \bar{\lambda}$, otherwise the period ΔT will be too large. The film thickness is bounded from above by the usual condition for observing the quantum size effect, since the amplitude of the oscillations decreases exponentially with increasing L.

The foregoing analysis of the oscillations of the thermodynamic quantities is valid near the edge of a nondegenerate band, for example for holes in bismuth, when the quadratic dispersion law is valid. For a more complicated dispersion law, for example for electrons in bismuth (see the next section), one obtains accordingly a more complicated temperature dependence of the amplitude and phase of the oscillations.

In the case of an arbitrary dispersion law, the oscillations of the thermodynamic quantities can be calculated in the quasiclassical approximation using the quantized energy spectrum $\epsilon_n(p)$ of the conduction electron in the film, determined by the condition[8] for the quantization of the chord of the equal-energy surface:

$$P_z \equiv |p_{z1}-p_{z2}| = 2\pi\hbar n/L, \quad n=1,2,3,\ldots \tag{8}$$

In this case we obtain from (1) for Ω_{osc}

$$\Omega_{\text{osc}}=\frac{2VT}{\pi\hbar L^2}\sum_{k,\,s=1}^{\infty}\frac{1}{k^2}\operatorname{Re}\Big\{(\pm J_s)^{-1/2}$$
$$\times \exp i\left[k\frac{L}{\hbar}P_z(\zeta+i\pi(2s-1)T)\pm\frac{\pi}{4}\pm\frac{\pi}{4}\right]\Big\}, \tag{9}$$

where the quantity

$$J_s=\frac{\partial^2 P_z}{\partial p_x^{\,2}}\frac{\partial^2 P_z}{\partial p_y^{\,2}}-\left(\frac{\partial^2 P_z}{\partial p_x\,\partial p_y}\right)^2$$

is evaluated at the energy $\epsilon = \zeta + i\pi(2s - 1)T$. Equal signs, which coincide with the sign of $\partial^2 P_Z/\partial P_X^2$, are chosen in the phase of the oscillations if $J > 0$ and $T = 0$, and opposite signs are chosen in the case when $J < 0$; $(\pm J_S)^{-1/2} = |J|^{-1/2}$ when $T = 0$.

The amplitude and the phase of the oscillations in (9) are determined by the dependence of the extremal chord P_Z $(\partial P_Z/\partial p_X = \partial P_Z/\partial p_y = 0)$ on the complex argument $\zeta + i\pi(2s - 1)T$. At not too high temperatures, it may be convenient to use the expansion

$$P_z(\zeta+i\pi(2s-1)T)=\sum_{\nu=0}^{\infty}\frac{1}{\nu!}\frac{d^\nu P_z}{d\zeta^\nu}(i\pi(2s-1)T)^\nu. \tag{10}$$

At temperatures $\pi T/\zeta \ll 1$, only the first terms of the expansion (10) are significant for the oscillations (9); these terms satisfy the condition

$$\frac{L}{\hbar}\frac{d^\nu P_z}{d\zeta^\nu}(\pi T)^\nu \gtrsim 1. \tag{10$'$}$$

For not too thin films it is possible to retain in (9) only one term with $k = s = 1$. At temperatures

$$\hbar/LP_z \lesssim (\pi T/\zeta)^2 \ll 1 \tag{10$''$}$$

we obtain for Ω_{osc} from (9)

$$\Omega_{\text{osc}}=\frac{2VT}{\pi\hbar L^2}|J|^{-1/2}\exp\left[-\pi T\frac{L}{\hbar}\left(\frac{1}{v_{z1}}+\frac{1}{|v_{z2}|}\right)\right]$$
$$\times\cos\left\{\frac{L}{\hbar}\left[P_z(\zeta)+\frac{\pi^2T^2}{2}\left(\frac{1}{m_{z1}^{\;*}v_{z1}^{\;3}}+\frac{1}{m_{z2}^{\;*}|v_{z2}|^3}\right)\right]\pm\frac{\pi}{4}\pm\frac{\pi}{4}\right\}. \tag{11}$$

Here v_{Zi} and m^*_{Zi} are respectively the values of the velocity $v_Z = \partial E/\partial p_Z$ and of the effective mass $m_Z^{*-1} = \partial^2 E/\partial p_Z^2$ at the points where the Fermi surface intersects the extremal chord. These quantities can be determined from an investigation of the temperature dependence of the phase and of the amplitude of the oscillations in the given temperature interval (10″). With increasing temperature, it is necessary to take into account higher terms of the expansion (10).

At low temperatures, $(LP_Z/\hbar)(\pi T/\zeta)^2 \ll 1$, we can neglect the temperature dependence of the phase of the oscillation. In this case the period ΔL of the size-effect oscillations does not depend on T and is determined by the extremal chord $P_Z(\zeta)$[9].

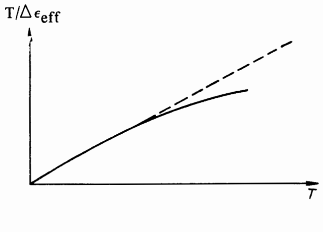

FIG. 1. Temperature dependence of the quantity $T/\Delta\epsilon_{\text{eff}}$, which determines the amplitude A of the quantum size-effect oscilations ($A \sim \exp(-2\pi^2 T/\Delta\epsilon_{\text{eff}})$), in the case of quadratic dispersion.

3. SEMIMETALS. TWO-BAND MODEL

Let us consider in somewhat greater detail the oscillations of the thermodynamic quantities of the electrons in bismuth, for which the quadratic-dispersion approximation does not hold. This is of interest in itself because of experimental investigations of the QSE on bismuth films. In addition, the analysis presented here for the size-effect oscillations in the approximation of two-band model[10] shows that the temperature dependence of the amplitude and of the phase of the oscillations is sensitive to the dispersion law, and its investigation may yield valuable information concerning the structure of the electronic energy spectrum.

In the two-band approximation[10] (see the review[11]) we have for the energy spectrum of the electrons in bismuth

$$\varepsilon\left(1+\frac{\varepsilon}{\varepsilon_g}\right)=\frac{1}{2}\sum_{i,\,k=1}^{3}\alpha_{ik}^{-1}p_ip_k, \tag{12}$$

where ϵ_g is the gap between the conduction and valence bands.

Using the quantization condition (8), we obtain from (12) for the quantum energy levels in the quasiclassical approximation

$$\varepsilon_n(\mathbf{p})\left(1+\frac{\varepsilon_n(\mathbf{p})}{\varepsilon_g}\right)=\frac{1}{2m_z}\left(\frac{\pi\hbar n}{L}\right)^2+\frac{p_x^2}{2m_x}+\frac{p_y^2}{2m_y}, \tag{13}$$

where the effective masses m_z, m_x, and m_y and the directions of the x and y axes are expressed in a definite manner in terms of the components of the tensor α_{ik}^{-1}.

We shall need in what follows the mass m_z, which, following[12], can be expressed in terms of the principal values m_1, m_2, and m_3, of the tensor α_{ik}:

$$\begin{aligned} m_{z\nu}^{-1}&=\frac{1}{m_1}\sin^2\vartheta\cos^2\left(\varphi+\frac{2\pi\nu}{3}\right)+\frac{1}{m_2}\left(\sin\alpha\cos\vartheta\right.\\ &\left.+\cos\alpha\sin\vartheta\sin\left(\varphi+\frac{2\pi\nu}{3}\right)\right)^2+\frac{1}{m_3}\left(\cos\alpha\cos\vartheta\right.\\ &\left.-\sin\alpha\sin\vartheta\sin\left(\varphi+\frac{2\pi\nu}{3}\right)\right)^2, \qquad \nu=0,1,2, \end{aligned} \tag{13'}$$

where α is the angle of inclination of the principal ellipsoid to the basal plane, ϑ and φ are angles determining the direction of the normal to the film relative to the crystallographic axes, and the index ν numbers the electron ellipsoids.

As a result of analogous calculations we obtain for the oscillating part Ω_{osc} of the thermodynamic potential of the electrons in bismuth films

$$\Omega_{\mathrm{osc}}=\sum_{\nu=0}^{2}\Omega_{\mathrm{osc}}^{(\nu)};$$

$$\begin{aligned}\Omega_{\mathrm{osc}}^{(\nu)}&=\frac{VT}{\pi\hbar L^2}\frac{\sqrt{m_1m_2m_3}}{m_{z\nu}^{3/2}}P_{z\nu}(\zeta)\sum_{k,\,s=1}^{\infty}\frac{1}{k^2}\exp\left(-k\frac{L}{\hbar}P_{z\nu}\beta_s(T)\right)\\ &\times\left[\gamma_s(T)\sin\left(k\frac{L}{\hbar}P_{z\nu}\gamma_s(T)\right)+\beta_s(T)\cos\left(k\frac{L}{\hbar}P_{z\nu}\gamma_s(T)\right)\right],\end{aligned} \tag{14}$$

where the extremal chord $P_{z\nu}$ is determined by the formula

$$P_{z\nu}=2\sqrt{2m_{z\nu}\zeta(1+\zeta/\varepsilon_g)}, \tag{15}$$

ζ is the chemical potential reckoned from the bottom of the conduction band,

$$\begin{aligned}\gamma_s&=\frac{1}{\sqrt{2}}\{\sqrt{(1+\Theta_s^2)(1+\Theta_{sg}^2)}+1-\Theta_s\Theta_{sg}\}^{1/2},\\ \beta_s&=\frac{1}{\sqrt{2}}\{\sqrt{(1+\Theta_s^2)(1+\Theta_{sg}^2)}-1+\Theta_s\Theta_{sg}\}^{1/2};\end{aligned} \tag{16}$$

Θ_s is defined by formula (3′) and

$$\Theta_{sg}=\pi(2s-1)T/(\zeta+\varepsilon_g). \tag{16'}$$

Summing (14) over s, we obtain for $\Omega_{\mathrm{osc}}^{(\nu)}$ at low temperatures $(\pi T/\zeta)^2\ll 1$ the following periodic oscillating function

$$\begin{aligned}\Omega_{\mathrm{osc}}^{(\nu)}&=\frac{VT}{\sqrt{2}\pi\hbar L^2}\frac{\sqrt{m_1m_2m_3}}{m_{z\nu}^{3/2}}P_{z\nu}\\ &\times\sum_{k=1}^{\infty}\frac{1}{k^2}\sin\left(k\frac{L}{\hbar}P_{z\nu}\right)\Big/\operatorname{sh}\left(k\frac{L}{\hbar}P_{z\nu}\frac{\pi T}{2\zeta}\frac{2\zeta+\varepsilon_g}{\zeta+\varepsilon_g}\right)\end{aligned} \tag{17}$$

with a period

$$\Delta L_\nu^{(1)}=2\pi\hbar/P_{z\nu}. \tag{17'}$$

At not too low temperatures, only one harmonic with k = 1 is significant in (17), and we have for $\Omega_{\mathrm{osc}}^{(\nu)}$ in this case

$$\begin{aligned}\Omega_{\mathrm{osc}}^{(\nu)}&=\frac{\sqrt{2}VT}{\pi\hbar L^2}\frac{\sqrt{m_1m_2m_3}}{m_{z\nu}^{3/2}}P_{z\nu}\sin\left(\frac{L}{\hbar}P_{z\nu}\right)\\ &\times\exp\left(-\frac{L}{\hbar}P_{z\nu}\frac{\pi T}{2\zeta}\frac{2\zeta+\varepsilon_g}{\zeta+\varepsilon_g}\right).\end{aligned} \tag{18}$$

At high temperatures, $\pi T\gg\zeta$, ϵ_g, we obtain from (14)

$$\begin{aligned}\Omega_{\mathrm{osc}}^{(\nu)}&=\frac{\sqrt{2}\pi VT^3}{\hbar L^2\zeta(\zeta+\varepsilon_g)}\frac{\sqrt{m_1m_2m_3}}{m_{z\nu}^{3/2}}P_{z\nu}\cos\left(\frac{L}{\hbar}P_{z\nu}\frac{2\zeta+\varepsilon_g}{2\sqrt{\zeta(\zeta+\varepsilon_g)}}\right)\\ &\times\exp\left(-\frac{L}{\hbar}P_{z\nu}\frac{\pi T}{\sqrt{\zeta(\zeta+\varepsilon_g)}}\right),\end{aligned} \tag{19}$$

which oscillates with a period

$$\Delta L_\nu^{(2)}=\frac{2\pi\hbar}{P_{z\nu}}\frac{\sqrt{1+\varepsilon_g/\zeta}}{1+\varepsilon_g/2\zeta}. \tag{19'}$$

The oscillation amplitude is proportional to $\exp(-2\pi^2T/\Delta\epsilon_{\mathrm{eff}})$, where

$$\Delta\varepsilon_{\mathrm{eff}}=\begin{cases}\dfrac{\pi\hbar}{L}\sqrt{\dfrac{\zeta}{2m_{z\nu}}}\dfrac{\sqrt{1+\zeta/\varepsilon_g}}{1/2+\zeta/\varepsilon_g}, & \left(\dfrac{\pi T}{\zeta}\right)^2\ll 1\\[2ex] \dfrac{\pi\hbar}{L}\sqrt{\dfrac{\varepsilon_g}{2m_{z\nu}}}, & \pi T\gg\zeta,\ \varepsilon_g\end{cases}. \tag{20}$$

Thus, in the limit of low and high temperatures, the phase of the size-effect oscillations of the thermodynamic quantities does not depend on the temperature. The values of the oscillation period ΔL_ν and the values of $\Delta\epsilon_{\mathrm{eff}}$ are different in the temperature regions under consideration and are likewise independent of T.

At low temperatures, $(\pi T/\zeta)^2\ll 1$, only the energies near the chemical potential ζ are of importance, so that in the vicinity of ζ the energy spectrum can be regarded with sufficient accuracy as equidistant, with a level spacing $\Delta\epsilon_\nu^{(1)}$. In the region of high temperatures, $\pi T\gg\zeta$, ϵ_g, the energies $\epsilon_n\gg\epsilon_g$ are significant, and it follows from (13) that in this case the energy spectrum is equidistant with level spacing $\Delta\epsilon_\nu^{(2)}$. The extremal values of $\Delta\epsilon_\nu^{(1)}$ and $\Delta\epsilon_\nu^{(2)}$ are determined from (13) with $p_x=p_y=0$ respectively,

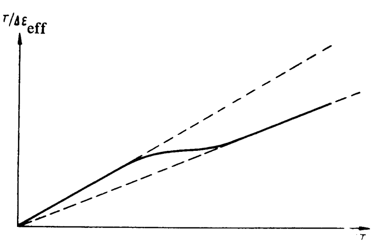

FIG. 2. Temperature dependence of $T/\Delta\epsilon_{\text{eff}}$ for electrons in bismuth, in the two-band-model approximation.

but for $\epsilon_n \to \infty$ and $\epsilon_n \gg \epsilon_g$. In accordance with (20), the values of $\Delta\epsilon_{\text{eff}}$ coincide with $\Delta\epsilon_\nu^{(1)}$ and $\Delta\epsilon_\nu^{(2)}$ in the corresponding temperature intervals. It turns out that $\Delta\epsilon_\nu^{(2)} > \Delta\epsilon_\nu^{(1)}$, and this is of importance in the estimate of the oscillation amplitude.

From the measurement of $\Delta\epsilon_{\text{eff}}$ or of the oscillation periods ΔL_ν in the limit of high and low temperatures it is possible to find the ratio ϵ_g/ζ. In this case, if the effective mass $m_{z\nu}$ on the bottom of the conduction band is known, it is possible to determine the values of the energy gap ϵ_g and of the chemical potential ζ.

In the temperature region $\pi T \sim \zeta$, ϵ_g, a transition takes place from one oscillation regime to another. An important role is played here by the fact that the energy spectrum has a non-equidistant character, as revealed by the temperature dependence of $\Delta\epsilon_{\text{eff}}$ and of the oscillation phase (see Fig. 2).

4. MAGNETIC SUSCEPTIBILITY

Investigations of the quantum oscillations of the magnetic susceptibility of films in perpendicular magnetic fields is important[13,14] in the study of the Fermi surface. Additional information concerning the electronic energy spectrum of the films can be obtained by investigating the temperature dependence of the amplitude and of the phase of the quantum oscillations of the thermodynamic quantities with changing magnetic field at not too low temperatures.

The thermodynamic potential Ω of the conduction electrons in a film in a perpendicular magnetic field is given by

$$\Omega = -T\frac{VeH}{2\pi\hbar cL}\sum_{\sigma=1}^{2}\sum_{n,\nu}\ln\left(1+\exp\frac{\zeta_\sigma-\varepsilon_{n,\nu+1/2}}{T}\right),$$

$$\zeta_\sigma = \zeta + (-1)^\sigma \hbar eH/2m_0c. \tag{21}$$

In weak magnetic fields, $\Delta\epsilon_H \ll \Delta\epsilon_L$ ($\Delta\epsilon_H$ is the distance between the Landau levels and $\Delta\epsilon_L$ is the distance between the size-quantized energy levels), the quantum energy spectrum $\epsilon_{n,\nu+1/2}$ is determined in the quasiclassical approximation in the magnetic field by the quantization condition[14]

$$S_n(\varepsilon) = \frac{2\pi\hbar eH}{c}\left(\nu+\frac{1}{2}\right). \tag{22}$$

Here $S_n(\epsilon)$ is the area bounded by the contour $\epsilon_n(p) = \epsilon$, where $\epsilon_n(p)$ is the size-quantized spectrum at $H = 0$. The condition (22) is the analog of the condition obtained by I. Lifshitz[15] and by Onsager[16] for the quantization of the energy spectrum of an electron with an arbitrary dispersion law in a bulky sample in a magnetic field.

Following[17], let us calculate the sum over ν in (21) by using the Poisson formula. As a result we obtain for the oscillating part of the thermodynamic potential

$$\Omega_{\text{osc}} = \frac{VTeH}{\pi\hbar Lc}\sum_{\sigma=1}^{2}\sum_{n=1}^{\infty}\sum_{k,\,s=1}^{\infty}\frac{(-1)^k}{k}\operatorname{Re}\exp\left[\frac{ikc}{\hbar eH}S_n(\zeta_\sigma+i\pi(2s-1)T)\right]. \tag{23}$$

In the considered region of magnetic fields and temperatures, only terms with $k = s = 1$ are significant in (23), and we can use the expansion of $S_n(\zeta_\sigma + i\pi T)$ in powers of T. For the oscillating part of the magnetic moment $M_{z.\text{osc}} = -\partial\Omega_{\text{osc}}/\partial H$ we thus obtain

$$M_{z\,\text{osc}} = \frac{VT}{\pi\hbar^2 HL}\sum_\sigma\sum_n S_n(\zeta_\sigma)$$
$$\times\exp\left[-2\pi^2\frac{cT}{\hbar eH}\sum_{l=0}^{\infty}\frac{(-1)^l}{(2l+1)!}(\pi T)^{2l}\frac{d^{2l}m_n^*}{d\zeta_\sigma^{2l}}\right]$$
$$\times\sin\left\{\frac{c}{\hbar eH}\left[S_n(\zeta_\sigma)+2\pi\sum_{l=1}^{\infty}\frac{(-1)^l}{(2l)!}(\pi T)^{2l}\frac{d^{2l-1}m_n^*}{d\zeta_\sigma^{2l-1}}\right]\right\}, \tag{24}$$

where $m_n^* = dS_n(\zeta)/2\pi d\zeta$ is the effective mass corresponding to the given subband $\epsilon_n(p) = \zeta$.

When $(\pi T/\zeta_n)^2 \ll 1$, the only terms that are significant in the series in terms of l in (24) are those satisfying the condition $(cS_n/\hbar eH)(\pi T/\zeta_n)^2 \gtrsim 1$, where ζ_n is the chemical potential reckoned from the edge of the corresponding subband.

The number of terms in the sum over n in (24) is determined by the condition $S_n(\zeta_\sigma) \geq 0$. For thin films near the "semimetal–dielectric" transition[18] due to the size quantization, this number is small. The opposite case, when a large number of terms in the sum over n is important in (21), is considered in[14] [2]).

Differentiating $M_{z.\text{osc}}$, we obtain in the temperature interval

$$\hbar eH/cS_n \lesssim (\pi T/\zeta_n)^2 \ll 1 \tag{25}$$

the following expression for the oscillations of the magnetic susceptibility χ_{osc}:

$$\chi_{\text{osc}} = -\frac{2VTc}{\pi\hbar^3 LeH^3}\sum_n S_n^2(\zeta)\cos\left\{\frac{c}{\hbar eH}\left[S_n(\zeta)-\pi^3T^2\frac{dm_n^*}{d\zeta}\right]\right\}$$
$$\times\cos\left(\pi\frac{m_n^*}{m_0}\right)\exp\left(-2\pi^2\frac{cm_n^*T}{\hbar eH}\right). \tag{26}$$

As usual, $S_n(\zeta_\sigma)$ has been expanded in powers of H and the principal terms were retained upon differentiation.

In the case when the effective mass m_n^* depends on the energy, as is typical of a non-equidistant energy spectrum, the phase of the magnetic-susceptibility oscillations turns out to depend on the temperature[3]).

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[2]) The magnetic properties of metallic films in the case of a quadratic dispersion law in a perpendicular magnetic field were considered in [19] for $T \ll \zeta$.

[3]) Formulas (23)–(26) make it possible to take into account the dependence of ζ on the temperature. For electrons and holes in a semimetal, the change of the chemical potential is limited by the electron neutrality condition, so that $\pi T/\zeta \gtrsim 1$ is satisfied in a wide temperature interval, and the non-equidistant character of the energy spectrum turns out to be significant in the temperature dependence of the amplitude and phase of the quantum oscillations of the thermodynamic (and kinetic) quantities.

Observation of this effect calls for sufficiently thin films, in which ζ_n is not too large ($\zeta_n \to 0$ as $L \to L_{cr}$, where L_{cr} is the thickness of the semimetallic film at which the "semimetal—dielectric" transition takes place). In this case the condition (25) is satisfied in magnetic fields and at temperatures at which observation of quantum oscillations of the magnetic susceptibility is possible.

We have confined ourselves here to an examination of quantum oscillations of the thermodynamic quantities. Similar features at high temperatures are possessed also by the quantum oscillations of the kinetic quantities.

In conclusion, I am deeply grateful to M. I. Kaganov for interest in the work and for valuable remarks. I am indebted to E. I. Bukhshtab and Yu. F. Komnik for a discussion of the experimental data on quantum size effects at high temperatures.

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[1] I. M. Lifshitz and A. M. Kosevich, Izv. Akad. Nauk SSSR, seriya fiz. 19, 395 (1955).

[2] V. B. Sandomirskiĭ, Zh. Eksp. Teor. Fiz. 52, 158 (1967) [Sov. Phys.-JETP 25, 101 (1967)].

[3] Yu. F. Ogrin, V. N. Lutskiĭ and M. I. Elinson, ZhETF Pis. Red. 3, 114 (1966) [JETP Lett. 3, 71 (1966)].

[4] Yu. F. Komnik and E. I. Bukhshtab, ZhETF Pis. Red. 6, 536 (1967) [JETP Lett. 3, 71 (1966)].

[5] Yu. F. Ogrin, V. N. Lutskiĭ, M. U. Arifova, V. I. Kovalev, V. B. Sandomirskiĭ and M. I. Elinson, Zh. Eksp. Teor. Fiz. 53, 1218 (1967) [Sov. Phys.-JETP 26, 714 (1968)].

[6] H. A. Combet and J. Y. LeTraon, Sol. State. Com., 6, 85 (1968).

[7] F. I. Itskovich, Izv. Vuzov, Fizika, 2, 13 (1961).

[8] S. S. Nedorezov, Zh. Eksp. Teor. Fiz. 51, 868 (1966) [Sov. Phys.-JETP 24, 578 (1967)].

[9] I. O. Kulik, ZhETF Pis. Red. 5, 652 (1967) [JETP Lett. 5, 143 (1967)].

[10] B. Lax, Bull. Am. Phys. Soc., 5, 167 (1960).

[11] L. A. Fal'kovskiĭ, Usp. Fiz. Nauk, 94, 3 (1968) [Sov. Phys.-Usp. 11, 1 (1968)].

[12] M. I. Kaganov, S. S. Nedorezov and A. M. Rustamova, Paper at Soviet-Japanese Conference on Low-temperature Physics, Novosibirsk, 1969; Fiz. Tverd. Tela 12, No. 8 (1970) [Sov. Phys.-Solid State 12, No. 8 (1971)].

[13] V. N. Lutskii and E. P. Fesenko, Fiz. Tverd. Tela 10, 3661 (1968) [Sov. Phys.-Solid State 10, 2902 (1969)].

[14] S. S. Nedorezov, Zh. Eksp. Teor. Fiz. 57, 907 (1969) [Sov. Phys.-JETP 30, 495 (1970)].

[15] I. M. Lifshitz and M. I. Kaganov, Usp. Fiz. Nauk 69, 419 (1959) [Sov. Phys.-Usp. 2, 831 (1959)].

[16] L. Onsager, Phil. Mag., 43, 1006 (1952).

[17] I. M. Lifshitz and A. M. Kosevich, Zh. Eksp. Teor. Fiz. 29, 730 (1955) [Sov. Phys.-JETP 29, 636 (1956)].

[18] V. N. Lutskiĭ, ZhETF Pis. Red. 2, 391 (1965) [JETP Lett. 2, 245 (1965)].

[19] L. E. Gurevich and A. Ya. Shik, Zh. Eksp. Teor. Fiz. 54, 1873 (1968) [Sov. Phys.-JETP 27, 1006 (1968)].

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