

ELECTRONIC PROPERTIES OF CONDUCTING SYSTEMS

Thermoelectric effect in layered conductors at low temperatures

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The linear response of the electronic system of a layered conductor to the presence of a temperature gradient is investigated theoretically. The dependence of the thermoelectric power on the temperature and external magnetic field is found at temperatures below the Debye temperature. Experimental investigation of this dependence will make it possible to study different relaxation mechanisms in a system of conduction electrons and to determine the structure of the electronic energy spectrum. © 2009 American Institute of Physics. [doi:10.1063/1.3253405]

In a conductor the electron current density $\mathbf{j}$, the temperature gradient ∇T , and the electric field $\mathbf{E}$ are interrelated by the following linear relation:

$$j_i = \sigma_{ij} E_j - \alpha_{ij} \frac{\partial T}{\partial x_j}. \quad (1)$$

A thermoelectric field inevitably arises in a nonuniformly heated sample even in the absence of current-conducting contacts ($\mathbf{j}=0$):

$$E_i = \rho_{il} \alpha_{lj} \frac{\partial T}{\partial x_j}, \quad (2)$$

where ρ_{ij} is the electric-resistivity tensor, which is inverse to the electric-conductivity tensor σ_{ij} .

Investigation of the thermoelectric effect in layered conductors in a strong magnetic field $\mathbf{B}$ yields detailed information on their electric energy spectrum.^{1,2} No less important information on charge carriers can be obtained by studying the temperature dependence of the thermoelectric field. This is because the kinetic coefficients

$$\sigma_{ik} = \frac{2e^3 B}{c(2\pi\hbar)^3} \int \frac{\partial f_0(\varepsilon)}{\partial \varepsilon} d\varepsilon \int dp_B \times \int_{-\infty}^0 \exp(t/\tau_p) dt \int_0^{T_B} dt' v_i(t') v_k(t' + t), \quad (3)$$

$$\alpha_{ik} = \frac{2e^2 B}{c(2\pi\hbar)^3} \int \frac{\partial f_0(\varepsilon)}{\partial \varepsilon} \frac{\varepsilon - \mu}{T} d\varepsilon \int dp_B \times \int_{-\infty}^0 \exp(t/\tau_\varepsilon) dt \int_0^{T_B} dt' v_i(t') v_k(t' + t), \quad (4)$$

obtained by solving Boltzmann's kinetic equation describe different relaxation processes in the system of charge carriers. The components of the tensor σ_{ik} are related with the electron momentum relaxation, which is characterized by the

time τ_p , and the components of the tensor α_{ik} depend on the energy relaxation time τ_ε . Here e , $\mathbf{v}$, p_B , and ε are the charge, velocity, projection of the momentum on the direction of the magnetic field, and the energy of the conduction electrons, $f_0(\varepsilon)$ is the equilibrium Fermi distribution function, μ is the chemical potential of the system of electrons, c is the speed of light, $\hbar$ is Planck's constant, t is the time during which a charge moves in a magnetic field according to the equation

$$\frac{\partial \mathbf{p}}{\partial t} = \frac{e}{c} [\mathbf{v} \mathbf{B}].$$

When the motion of a charge in a magnetic field is periodic the quantity T_B is the period of the motion. However, if an electron undergoes aperiodic motion on an open trajectory in momentum space, then T_B is the characteristic time required for the electron to traverse one period of the reciprocal lattice. Strictly speaking, in this case it is necessary to average over a large section that an electron traverses on the open trajectory over a time of the order of its free flight time.³

In the absence of an external magnetic field the intensity of the thermoelectric field

$$E_i = \frac{\pi^2}{3e} \left(\frac{T}{\mu} \right) Q_{ik} \frac{\tau_\varepsilon}{\tau_p} \frac{\partial T}{\partial x_k} \quad (5)$$

is proportional to the ratio of the relaxation times τ_ε and τ_p . Here Q_{ik} is a dimensionless tensor which is temperature-independent.

We shall now examine the thermoelectric effect in a quasi-two-dimensional conductor at temperatures much less than the Debye temperature T_D , when the temperature dependence of the relaxation time τ_ε is much different from that of τ_p . At temperatures close to zero the electron system in degenerate conductors relaxes primarily as a result of scattering of charge carriers by impurity centers and other crystal defects. In this case the momentum and energy relaxation times

τ_p and τ_e are temperature-independent. As temperature increases, an additional charge-carrier relaxation mechanism is actuated—scattering by vibrations of the crystal lattice. According to Matthiessen's rule, each scattering mechanism makes an additive contribution to the relaxation process and

$$\frac{1}{\tau} = \frac{1}{\tau^{(im)}} + \frac{1}{\tau^{(eph)}}, \quad (6)$$

where $1/\tau^{(im)}$ is the charge-carrier-phonon collision frequency. For $T \ll T_D$, because of the electron-phonon scattering angles are small a much larger number of collisions with phonons are needed for momentum relaxation than for energy relaxation (see, for example, the monograph Ref. 4). As a result, τ_e decreases much more rapidly with increasing temperature than does τ_p . The temperature dependence of τ_e has the form

$$\tau_e = \left\{ \frac{1}{\tau^{(im)}} + \frac{1}{\tau_0} \tilde{T}^n \right\}^{-1}, \quad (7)$$

where $\tilde{T} = T/T_D$, τ_0 is the time characterizing the energy relaxation of electrons as a result of electron collisions with phonons at the Debye temperature, and the dimensionality of the system determines the n in the exponent. For a three-dimensional metal $n=3$, and for a two-dimensional conductor $n=2$.⁵

At low temperatures, when the momentum relaxation of the electrons is mainly due to electron collisions with impurity centers, i.e. $\tau_p = \tau^{(im)}$, and electron-phonon scattering processes make an appreciable contribution to the energy relaxation, the temperature dependence of the thermoelectric field with $\mathbf{B}=0$ can be represented in the form

$$E_i = \frac{\pi^2 T_D}{3e\mu} Q_{ik} f(\tilde{T}) \frac{\partial T}{\partial x_k}, \quad f(\tilde{T}) = \frac{\tilde{T}}{(\tau^{(im)}/\tau_0) \tilde{T}^n}. \quad (8)$$

In sufficiently pure semiconductors ($\tau^{(im)} \gg \tau_0$) the competition between different scattering mechanisms leads to the appearance of a maximum in the plot of the thermoelectric field versus temperature. For $\tilde{T} \ll (\tau_0/\tau^{(im)})^{1/n}$ the field E_i is proportional to $\tilde{T}$, but as temperature increases the charge-carrier-phonon collisions frequency $1/\tau_e^{(eph)} = (1/\tau_0) \tilde{T}^n$ becomes comparable to the frequency of carrier collisions with impurity centers $1/\tau^{(im)}$ and the increase with temperature is replaced by a decrease proportional to $\tilde{T}^{(1-n)}$. The maximum is attained at

$$\tau_e^{(eph)} = (n-1) \tau^{(im)}.$$

A plot of the function $f(\tilde{T})$ for $n=3$ is displayed in Fig. 1.

In layered conductors with a quasi-two-dimensional electron energy spectrum the temperature dependence of the relaxation time $\tau_e^{(eph)}$ at temperatures much less than the overlap integral $t_\perp$ of the wave functions of electrons belonging to different layers is essentially the same as in three-dimensional conductors and $\tau_e^{(eph)}$ is proportional to T^{-3} , though the coefficient of proportionality depends on the magnitude of the anisotropy of the charge-carrier spectrum. As temperature increases and T becomes greater than or comparable to $t_\perp$, $\tau_e^{(eph)}$ decreases quadratically with temperature. In organic conductors the overlap integral $t_\perp$ is ap-

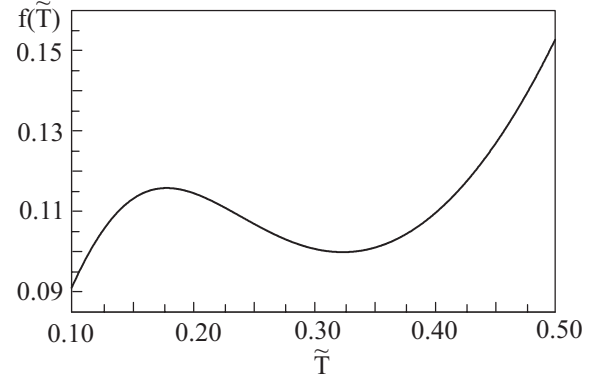


FIG. 1. Plot of the function $f(\tilde{T})$ for $n=3$ and $\tau^{(im)}/\tau_0=10^2$.

proximately 100 times less than the Fermi energy, i.e. comparable to the Debye temperature in order of magnitude; $\tau_e^{(eph)}$ is proportional to T^{-2} at temperatures close to T_D only in certain forms of graphite. Thus $\tau_e^{(eph)}$ is proportional to T^3 at all temperatures less than the Debye temperature T_D , and following Bloch's law we shall represent τ_p in the form

$$\frac{1}{\tau_p} = \frac{1}{\tau^{(im)}} + \frac{1}{\tau_0} \tilde{T}^5. \quad (9)$$

The energy spectrum of organic conductors is quite complicated, and in some of them several groups of charge carriers are responsible for electron transport while the Fermi surface consists of topologically different elements: weakly fluted cylinder and planes.^{6,7}

An external magnetic field affects differently the motion of charge carriers whose states belong to the weakly fluted cylinder or a fluted flat sheet of the Fermi surface. This is why the presence of such flat sheets of the Fermi surface is most easily revealed in a conductor placed in a magnetic field.

We shall examine as an example a conductor with two groups of charge carriers, which is placed in a strong magnetic field $\mathbf{B} = (0, B \sin \theta, B \cos \theta)$. We shall assume that the velocities $\pm \mathbf{v}_1$ of the electrons belonging to two flat sheets of the Fermi surface are predominantly oriented in a direction determined by an angle φ so that $v_{1x} = \pm v_1 \cos \varphi$ and $v_{1y} = \pm v_1 \sin \varphi$, and the dispersion law for the charge carriers belonging to a weakly fluted cylinder has the form

$$\varepsilon(\mathbf{p}) = \frac{p_x^2 + p_y^2}{2m} - 2t_\perp \cos \frac{ap_z}{\hbar}, \quad (10)$$

where $m = \text{const}$, a is distance between the layers, and $t_\perp$ can be less than the Fermi energy ε_F .

We shall assume that the angle θ of deviation of the magnetic field from the direction normal to the layers is not too close to $\pi/2$, so that all orbits of electrons with a quadratic dispersion law are closed and do not contain self-intersections. For $\cos \theta \gg mc/eB\tau$ the strong magnetic field condition $(T_B/\tau) \ll 1$ is automatically satisfied. The components of the conductor's kinetic coefficients σ_{ik} and α_{ik} are sums of the contributions of quasi-two-dimensional and quasi-one-dimensional charge carriers, which are easily calculated using the relations (3) and (4). Specifically, the following expression obtains for the electric-conductivity ten-

SOR:

$$\sigma_{ik} = \begin{pmatrix} \gamma^2 \sigma_2 - \gamma^2 \sigma_{zz} \tan^2 \theta + \sigma_1 \cos^2 \varphi & \gamma \sigma_2 - \gamma \sigma_{zz} \tan^2 \theta + \sigma_1 \cos \varphi \sin \varphi & -\gamma \sigma_{zz} \tan \theta \\ -\gamma \sigma_2 + \gamma \sigma_{zz} \tan^2 \theta + \sigma_1 \cos \varphi \sin \varphi & \gamma^2 \sigma_2 + \sigma_{zz} \tan^2 \theta + \sigma_1 \sin^2 \varphi & \sigma_{zz} \tan \theta \\ \gamma \sigma_{zz} \tan \theta & \sigma_{zz} \tan \theta & \sigma_{zz} \end{pmatrix}. \quad (11)$$

Here σ_1 and $\sigma_2 = (e^2 \varepsilon_F \tau_p / \pi \hbar^2 a)$ are the contributions to the electric conductivity along the layers with $\mathbf{B}=0$ of charge carriers whose states belong to a flat sheet of the Fermi surface and a fluted cylinder, $\gamma = mc / (eB \tau_p \cos \theta) = 1 / (\omega_c \tau_p) \ll 1$. Taking account of the contribution to σ_{zz} of charge carriers with a quasi-one-dimensional energy spectrum has no effect on the temperature dependence of the thermoelectric field. The component of the electric conductivity

$$\sigma_{zz} = \frac{2ae^2 m \tau_p t_{\perp}^2 \cos \theta}{\pi \hbar^4} J_0^2 \left(\frac{ap_F}{\hbar} \tan \theta \right) \equiv S_z \tau_p \quad (12)$$

in the leading-order approximation in the quasi-two-dimensionality parameter $t_{\perp} / \varepsilon_F = \eta \ll 1$ is quadratic in η and for $1 \ll \tan \theta \ll eB \tau_p / mc$ exhibits periodic oscillations as function of $\tan \theta$, where θ is the angle of deviation of the magnetic field from the normal to the layers. Here $J_n(x)$ is a Bessel function and p_F is the Fermi momentum.

Inverting the electric-conductivity tensor it is easily shown that for $\gamma \ll 1$ the following asymptotic expression holds for the resistivity tensor:

$$\rho_{ik} = \rho_0 \begin{pmatrix} 1 + q \frac{\sin^2 \varphi}{\gamma^2} & -\frac{1}{\gamma} - q \frac{\sin 2\varphi}{2\gamma^2} & \left(\frac{1}{\gamma} + q \frac{\sin 2\varphi}{2\gamma^2} \right) \tan \theta \\ \frac{1}{\gamma} - q \frac{\sin 2\varphi}{2\gamma^2} & 1 + q \frac{\cos^2 \varphi}{\gamma^2} & -q \frac{\cos^2 \varphi}{\gamma^2} \tan \theta \\ \left(-\frac{1}{\gamma} + q \frac{\sin 2\varphi}{2\gamma^2} \right) \tan \theta & -q \frac{\cos^2 \varphi}{\gamma^2} \tan \theta & \frac{1}{\sigma_{zz} \rho_0} + q \frac{\cos^2 \varphi}{\gamma^2} \tan^2 \theta \end{pmatrix}, \quad (13)$$

where $\rho_0 = 1 / (\sigma_1 + \sigma_2)$ and $q = \sigma_1 / \sigma_2$. The periodic variations of the component ρ_{zz} as a function of $\tan \theta$ are a manifestation of the particulars of the dispersion law for charge carriers on a sheet of the Fermi surface in the form of a fluted cylinder. Such oscillations have been observed in measurements of the magnetoresistance to current flow across the layers in a number of conductors of organic origin and have been studied quite thoroughly.⁸

The thermoelectric effect depends strongly on the presence of flat sheet of the Fermi surface, and different components of the thermoelectric field

$$E_i = \frac{\pi^2 T_D}{3e\mu} P_{ik}(T) \frac{\partial T}{\partial x_k} \quad (14)$$

exhibit very diverse behavior. For one group of charge carriers with a quasi-two-dimensional dispersion law the diagonal components of P_{ik} in the plane of the layers

$$P_{xx} = P_{yy} = \tilde{T} \quad (15)$$

in the leading-order approximation in the small parameters γ and η are much larger than the components

$$P_{xy} = -P_{yx} = \tilde{T} \left(\frac{1}{\omega_c \tau_p} - \frac{1-b}{\omega_c \tau_e} \right), \quad (16)$$

so that if the temperature gradient vector lies in the plane of the layers, then the thermoelectric field in this plane is directed mainly along ∇T and grows linearly with T . The components

$$P_{zx} = -\tilde{T} \left(\frac{1-b}{\omega_c \tau_e} - \frac{d}{\omega_c \tau_p} \right) \tan \theta, \quad (17)$$

$$P_{zy} = \tilde{T} \left(-1 + \frac{(d+b)\tau_e}{\tau_p} \right) \tan \theta \quad (18)$$

are different from zero only if the magnetic field deviates from the normal to the layers

$$P_{zz} = \tilde{T}(d+b), \quad (19)$$

and P_{xz} and P_{yz} are proportion to η^2 . Here $b = (\mu / \tau_e) \times (\partial \tau_e / \partial \mu) \equiv 1$ and the following expression holds for the coefficient $d = (\mu / S_z) (\partial S_z / \partial \mu)$:

$$d = -\frac{2\mu m a \tan \theta}{p_F \hbar} \frac{J_1 \left(\frac{ap_F}{\hbar} \tan \theta \right) J_0 \left(\frac{ap_F}{\hbar} \tan \theta \right)}{J_0^2 \left(\frac{ap_F}{\hbar} \tan \theta \right) + \phi_1 \gamma^2 + \phi_2 \eta^2}, \quad (20)$$

where ϕ_1 and ϕ_2 are of the order of 1 and take account of the corrections to S_z which were omitted in the leading-order approximation in the small parameters η and γ . As a result, the components P_{zk} of the thermoelectric field undergo gigantic oscillations. The function P_{zz} versus θ is displayed in Fig. 2.

The temperature dependence of the off-diagonal components of the tensor P_{ik} is determined by the temperature dependence of the relaxation times, and the relations (7) and (9) yield

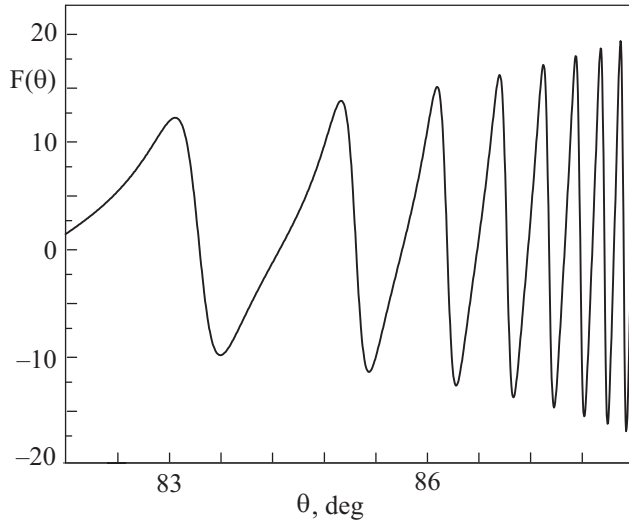


FIG. 2. Plot of the function $F(\theta)$, determining the function $P_{zz} = \tilde{T}F(\theta)$, versus the angle θ of deviation of the magnetic field from the normal to the layers in the case where there is only one group of charge carriers with a quasi-two-dimensional dispersion law.

$$P_{xy} = \frac{\tilde{T}}{\omega_c \tau^{(im)}} \left[b - (1-b) \frac{\tau^{(im)}}{\tau_0} \tilde{T}^3 + \frac{\tau^{(im)}}{\tau_0} \tilde{T}^5 \right], \quad (21)$$

$$P_{zx} = \frac{\tilde{T}}{\omega_c \tau^{(im)}} \left[d + b - 1 - (1-b) \frac{\tau^{(im)}}{\tau_0} \tilde{T}^3 + d \frac{\tau^{(im)}}{\tau_0} \tilde{T}^5 \right] \tan \theta, \quad (22)$$

$$P_{zy} = \tilde{T} \left[-1 + (d+b) \frac{1 + \frac{\tau^{(im)}}{\tau_0} \tilde{T}^5}{1 + \frac{\tau^{(im)}}{\tau_0} \tilde{T}^3} \right] \tan \theta. \quad (23)$$

In the presence of a quasi-one-dimensional group of charge carriers the components of the tensor P_{ik} contain terms which grow linearly with $\mathbf{B}$. These terms vanish for certain values of the angle φ and in all other cases they are the primary determinants of the thermoelectric effect in a strong magnetic field ($\omega\tau \gg 1$). If the temperature gradient vector lies in the plane of the layers, the diagonal components

$$P_{xx} = \rho_0 \tilde{T} \left[\sigma_2 + \sigma_1 \left((1-b) \frac{\tau_p}{\tau_\varepsilon} \sin^2 \varphi + a \frac{\tau_\varepsilon}{\tau_p} \cos^2 \varphi + \omega_c (\tau_p - a\tau_\varepsilon) \sin \varphi \cos \varphi \right) \right], \quad (24)$$

$$P_{yy} = \rho_0 \tilde{T} \left[\sigma_2 + \sigma_1 \left((1-b) \frac{\tau_p}{\tau_\varepsilon} \cos^2 \varphi + a \frac{\tau_\varepsilon}{\tau_p} \sin^2 \varphi - \omega_c (\tau_p - a\tau_\varepsilon) \sin \varphi \cos \varphi \right) \right] \quad (25)$$

grow linearly as functions of the magnetic field when both components v_{1x} and v_{2x} of the predominate direction of the velocity vector of electrons on a flat sheet of the Fermi surface are simultaneously different from zero. Then the tem-

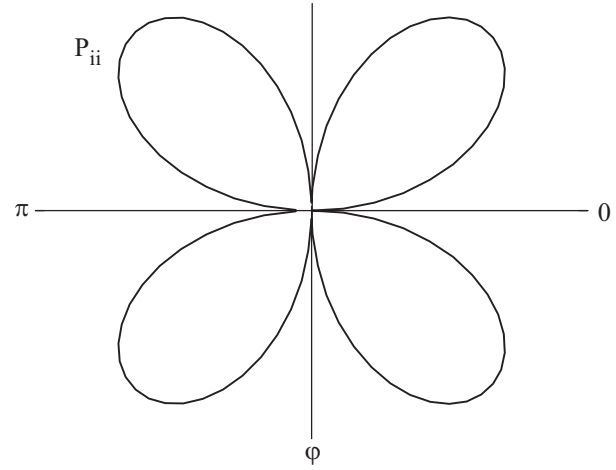


FIG. 3. P_{xx} and P_{yy} versus the angle φ (rotational diagram) for the case where two groups of charge carriers are present.

perature dependence of the components P_{xx} and P_{yy} in the leading-order approximation in the small parameter γ has the form

$$P_{xx} = -P_{yy} = \rho_0 \sigma_1 \omega_c \tau^{(im)} \tilde{T} \times \left(\frac{1}{1 + \frac{\tau^{(im)}}{x_0} \tilde{T}^5} - \frac{a}{1 + \frac{\tau^{(im)}}{x_0} \tilde{T}^3} \right) \sin \varphi \cos \varphi. \quad (26)$$

Here $a \equiv (\mu/\sigma_1)(\partial\sigma_1/\partial\mu) \cong 1$.

For the values of the angle φ such that $\sin 2\varphi = 0$, the terms P_{xx} and P_{yy} are independent of the magnetic field. The dependences of the diagonal component of the tensor P_{ik} on the angle φ and the temperature for the case of two groups of charge carriers are shown in Figs. 3 and 4.

The off-diagonal components

$$P_{yx} = -\rho_0 \tilde{T} \left[\sigma_2 \left(\frac{1}{\omega_c \tau_p} - \frac{1-b}{\omega_c \tau_\varepsilon} \right) - \sigma_1 \omega_c (\tau_p - a\tau_\varepsilon) \cos^2 \varphi \right], \quad (27)$$

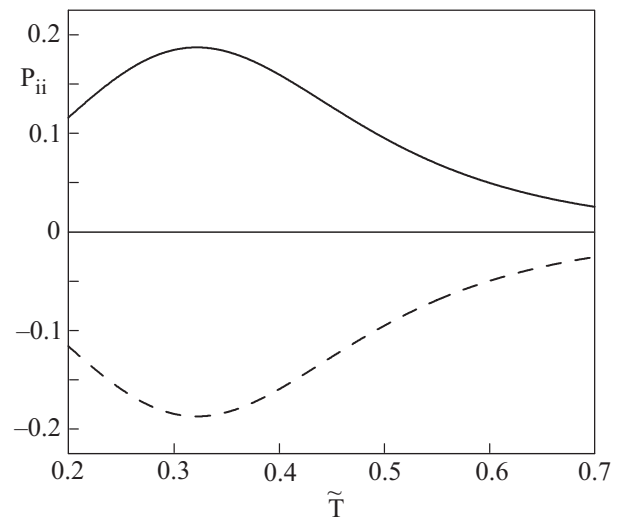


FIG. 4. Temperature dependence of P_{xx} (top curve) and P_{yy} (bottom curve) for $\sin 2\varphi \neq 0$ and $\tau^{(im)}/\tau_0 = 10^2$ in relative units for a two-band conductor.

$$P_{xy} = -\rho_0 \tilde{T} \left[\sigma_2 \left(\frac{1}{\omega_c \tau_p} - \frac{1-b}{\omega_c \tau_e} \right) - b_1 \omega_c (\tau_p - a \tau_e) \sin^2 \varphi \right], \quad (28)$$

$$P_{zx} = \rho_0 \tilde{T} \left[\sigma_2 \left(\frac{d}{\omega_c \tau_p} - \frac{1-b}{\omega_c \tau_e} \right) + \sigma_1 \left(\omega_c (\tau_p - a \tau_e) \cos^2 \varphi + \frac{d}{\omega_c \tau_p} \right) \right] \tan \theta, \quad (29)$$

$$P_{zy} = \rho_0 \tilde{T} \left[\sigma_2 \left(-1 + (d+b) \frac{\tau_e}{\tau_p} \right) + \sigma_1 \left(\omega_c (\tau_p - a \tau_e) \sin \varphi \cos \varphi + (d+b) \frac{\tau_e}{\tau_p} \right) \right] \tan \theta \quad (30)$$

likewise contain large terms which are proportional to γ^{-1} and whose temperature dependence is described by an expression similar to the relation (26).

It is easy to see from the relations presented above that the Nernst-Ettinghausen effect is clearly manifested when the temperature gradient is not orthogonal to the electron velocity vector $\mathbf{v}_1$.

The electric field in the direction of the normal to the layers increases with the angle θ of the deviation of the magnetic field from the normal to the layers and for $\tan \theta > 1$ it exceeds the electric field along the layers.

For $\tan \theta > eB\tau/mc = 1/\gamma_0$ there is no longer enough time for an electron to traverse over its free-flight time one complete revolution on a closed section of a sheet of the Fermi surface in the form of a weakly fluted cylinder. An electron moves along the normal to the layers by a small amount, and for $\theta = \pi/2$ its average velocity along the normal to the layers is zero. As a result, $\sigma_{zz} = \eta^2 \gamma_0^2 g \sigma_2$ in the leading-order approximation in the quasi-two-dimensionality parameter, where g is a dimensionless quantity of the order of 1. Then the components of the tensor P_{ik} which determine the thermoelectric field along the normal to the layers have the form

$$P_{zx} = \tilde{T} [\omega_c \tau_p - \rho_0 \sigma_2 (1+b) \omega_c \tau_e] \sin \varphi, \quad (31)$$

$$P_{zy} = -\tilde{T} [\omega_c \tau_p - (1+b) \omega_c \tau_e] \cos \varphi, \quad (32)$$

and the electric field grows linearly with increasing magnetic field. The components

$$P_{xx} = \rho_0 \tilde{T} [\sigma_2 (1+b) + \sigma_1 (a+b)] \frac{\tau_e}{\tau_p}, \quad (33)$$

$$P_{yy} = \tilde{T} (1+b) \frac{\tau_e}{\tau_p} \quad (34)$$

reach saturation in a strong magnetic field. The electric field along the layers is directed mainly along the temperature gradient, since the components

$$P_{yx} = P_{xy} (1+q) = \tilde{T} \eta^2 g \left[(1+b) \frac{\tau_e}{\tau_p} - 1 \right] \sin \varphi \cos \varphi \quad (35)$$

are proportional to η^2 and, just as the diagonal components, saturate for $\gamma_0 \ll 1$.

The presence of a group of charge carriers with a quasi-one-dimension dispersion law does not appreciably affect the thermoelectric effect when the magnetic field is almost parallel to the plane of the layers but it does change the entire picture of the behavior of the conductor in a strong magnetic field tilted away from the plane of the layers. The temperature and magnetic field dependences of the thermoelectric power are found to be very different for conductors with one quasi-two-dimensional group of charge carriers and for a conductor with a Fermi surface which also contains flat sheets. The diversity of these dependences gives rich material for studying the properties of charge carriers in low-dimensional conducting systems and makes it possible not only to reveal the presence of a flat sheet of the Fermi surface but also to determine the predominate direction of the velocities of electrons whose states belong to this sheet.

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