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Thermoelectric effect in layered conductors in a strong magnetic field*

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We have theoretically studied the thermoelectric effect in layered conductors with a quasi-two-dimensional electron energy spectrum of an arbitrary form in a quantizing magnetic field at low temperatures. Giant quantum oscillations of thermoelectric field versus the inverse magnetic field have been predicted, which will facilitate the experimental study of quantum oscillatory effects. Thermoelectric force in a layered conductor is shown to depend periodically upon the angle between the magnetic field direction and the normal to the layers. This orientation effect arises from the quasi-two-dimensional character of the charge carriers energy spectrum and is representative of layered conductors.

Key words: *thermoelectric field, layered conductor, quantum oscillations*

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Layered structures with sharply anisotropic metal type electrical conductivity have been intensively studied recently. The strong anisotropy of the kinetic coefficients of such conductors is attributed to the quasi-two-dimensional character of the energy spectrum for charge carriers. The dependence of the energy of conduction electrons on their momentum depends weakly on the momentum projection $p_z = \mathbf{pn}$ along the normal $\mathbf{n}$ to the layers and can be represented in the form of a rapidly converging series

$$\varepsilon(\mathbf{p}) = \sum_{k=0}^{\infty} \varepsilon_k(p_x, p_y) \cos\left(\frac{akp_z}{\hbar}\right). \tag{1}$$

Here a is the separation between layers, $\hbar$ is the Planck constant. The maximum values $\varepsilon_k^{\max}$ at the Fermi surface decrease significantly with increasing k so that $\varepsilon_1^{\max} = \eta\varepsilon_{\mathrm{F}} \ll \varepsilon_{\mathrm{F}}$ where the parameter of the quasi-two dimensionality η characterizes the anisotropy of the charge carriers spectrum.

In many-layered conductors of organic origin placed in a strong magnetic field $\mathbf{H}$ Shubnikov-de Haas oscillations have been observed [1]. This points to the presence of closed sections of the Fermi surface and to the fact that charge carriers mean lifetime τ is large enough and an electron can perform many rotations with frequency Ω during τ. We shall assume that the Fermi surface has the form of a weakly corrugated cylinder, which is in a good agreement with experimental investigations of galvanomagnetic effects and Shubnikov-de Haas oscillations in many tetrathiafulvalene-based complexes with charge transport.

In present paper we consider thermoelectric phenomena in layered conducting structures placed in a strong quantizing magnetic field $\mathbf{H} = (0, H\sin\vartheta, H\cos\vartheta)$. In the quasi-classical approximation, when the interval between quantized energy levels of charge carriers is much less than the Fermi

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energy ε_{F}, the electron energy spectrum can be determined making use of the rule of quantizing areas

$$S(\varepsilon, p_H) = \frac{2\pi\hbar eH}{c}\left(n + \frac{1}{2}\right), \tag{2}$$

where n are nonnegative integers, e is the electron charge, c is the velocity of light, $S(\varepsilon, p_H)$ is the area of the section of the isoenergetic cut by the plane $p_H = \mathbf{pH}/H = \mathrm{const}$.

The linear response of electron system to weak perturbation by electric field $\mathbf{E}$ and temperature gradient ∇T

$$j_i = \sigma_{ij} E_j - \alpha_{ij}\,\frac{\partial T}{\partial x_j}, \tag{3}$$

$$q_i = \beta_{ij} E_j - \kappa_{ij}\,\frac{\partial T}{\partial x_j} \tag{4}$$

should be determined by means of the solution of the kinetic equation for the statistical operator $\hat{f}$.

Represent statistical operator $\hat{f}$ in the form $\hat{f} = \hat{f}_0 + \hat{f}_1 + \hat{f}_2$ where $\hat{f}_0$ is the equilibrium operator whose diagonal matrix elements f_0^{nn} are equal to the Fermi distribution function $f_0\{\varepsilon_n(p_H)\}$, and operators $\hat{f}_1$ and $\hat{f}_2$ describe the weak perturbations of electron system by the electric field and the temperature gradient. Then the kinetic equation takes the form [2,3]:

$$\frac{i}{\hbar}(\varepsilon_n - \varepsilon_{n'}) f_1^{nn'} + \hat{W}_{nn'}\{\hat{f}_1\} = e\mathbf{E}\mathbf{v}_{nn'}\,\frac{f_0(\varepsilon_n) - f_0(\varepsilon_{n'})}{\varepsilon_n - \varepsilon_{n'}}, \tag{5}$$

$$\frac{i}{\hbar}(\varepsilon_n - \varepsilon_{n'}) f_2^{nn'} + \hat{W}_{nn'}\{\hat{f}_2\} = \mathbf{v}_{nn'}\;\nabla T\,\frac{\mu - \varepsilon_{n'}}{T}\,\frac{\partial f_0(\varepsilon_{n'})}{\partial \varepsilon_{n'}}, \tag{6}$$

where $\mathbf{v}_{nn'}$ are the matrix elements of the electron velocity operator $\hat{\mathbf{v}}$, $\hat{W}\{\hat{f}_1\}$ and $\hat{W}\{\hat{f}_2\}$ describe the momentum and energy relaxation of charge carriers, respectively. Under strong magnetic field conditions ($\Omega\tau \gg 1$), which can be realized at low temperatures, charge carriers are scattered mainly by impurities and crystal defects, and we may not consider the kinetic equation for nonlinear phonons. At low temperatures the dominant scattering mechanism is elastic scattering, and the collision operators can be taken into account in the τ-approximation. In this case the relaxation times τ_ε and τ_p are of the same order of magnitude, and if one does not distinguish them, the kinetic coefficients satisfy the Kelvin-Onsager relations and the Wiedemann-Franz formula.

At low temperature there exist giant oscillations of the thermoelectric field that are caused by quantization of arbitrary motion of charge carriers in layered conductors [4]. Consider thermoelectric field $\mathbf{E}$ induced by the temperature gradient oriented along the normal to the layers

$$E_i = \rho_{ij}\;\alpha_{jz}\frac{\partial T}{\partial z}\,. \tag{7}$$

Here ρ_{ij} is the resistivity tensor.

The asymptotic expression for the thermoelectric field E_x for $\eta \ll 1$ and $(\Omega\tau)^{-1} \ll 1$ is determined by non-diagonal components of the resistivity tensor. In the lowest order approximation with respect to the small parameter $1/\Omega\tau$ the components ρ_{xy} and ρ_{xz} are the same as in the classical consideration. The quantum oscillations of these components appear in the highest order terms in expansion in $1/\Omega\tau$ that thus can be neglected [2].

The oscillatory dependence of E_x upon $1/H$ is determined by the quantum oscillations of the thermoelectric coefficients α_{ik}.

In the quasi-classical approximation the component α_{ik} can be obtained by making use of the Poisson formula and replacing integration with respect to n by integration with respect to energy. Straightforward calculations yield the following asymptotic expression

$$\alpha_{yz} = \alpha_{zz}\tan\vartheta = \frac{2e}{(2\pi\hbar)^3}\sum_{k=-\infty}^{\infty}(-1)^k \int \mathrm{d}\varepsilon\;\frac{\mu - \varepsilon}{T}\;\frac{\partial f_0(\varepsilon)}{\partial \varepsilon}\;\tau_\varepsilon \times \int \mathrm{d}p_H\;\left\{\frac{\partial S}{\partial p_H}\right\}^2\;\frac{(\sin\theta)^2}{\partial S/\partial \varepsilon}\;\exp\left\{\frac{\mathrm{i}kcS(\varepsilon, p_H)}{eH\hbar}\right\}, \tag{8}$$

where μ is the chemical potential of electrons. Here we have taken into consideration that

$$\bar{v}_y = \bar{v}_z \tan\theta = \bar{v}_H \sin\theta = \frac{\partial S/\partial p_H}{\partial S/\partial\varepsilon}\sin\theta, \tag{9}$$

where $\bar{v}_H$ is the drift velocity of charge carriers along the magnetic field averaged over the states of electron orbit $\varepsilon = \text{const},\ p_H = \text{const}$.

When calculating α_{yz}, it is necessary to allow for quantum oscillations of the relaxation time τ that result from the summation over the electron states in the incoming term of the collision integral. The quantum oscillations of the scattering amplitude of electrons by impurities in layered conductors have been obtained in [5–8]. If the condition

$$T \ll \hbar\Omega \ll \eta\varepsilon_{\mathrm{F}} \tag{10}$$

is satisfied the relaxation time in the Born approximation is given by

$$\begin{aligned} \frac{1}{\tau_p(\varepsilon)} &= \frac{1}{\tau_\varepsilon(\varepsilon)} = \frac{1}{\tau_0}\left\{1 + \left(\frac{e\hbar H}{m^* c\varepsilon}\right)^{1/2} \sum_{\mathrm{e}} \left|\frac{\partial^2 S_{\mathrm{e}}}{\partial p_H^2}\right|^{-1/2} g\right\}, \\ g &= \sum_k a_k(-1)^k k^{-1/2}\,\frac{ku}{\sinh ku}\,\cos\left[\frac{kcS_{\mathrm{e}}}{e\hbar H} + \frac{\pi}{4}s\right]\cos\frac{\pi k m^*}{m}\,. \end{aligned} \tag{11}$$

Here a_k are the numerical coefficients depending on the form of the dispersion law for charge carriers, τ_0 is their mean free path time in the absence of a magnetic field,

$$m^* = \frac{1}{2\pi}\,\frac{\partial S}{\partial\varepsilon}$$

is the cyclotron effective mass of charge carriers, m is the free electron mass,

$$u = \frac{2\pi^2 T}{\hbar\Omega}$$

and

$$s = \mathrm{sign}\frac{\partial^2 S_{\mathrm{e}}}{\partial p_H^2}\,.$$

The main contribution into the oscillatory part of τ_{osc} is made by a small part of electrons near the extreme cross-sections S_{e} of the Fermi surface, and the summation over all sections S_{e} should be taken.

The oscillating part of α_{zz},

$$\begin{aligned} \alpha_{zz}^{\mathrm{osc}} &= \frac{2e}{(2\pi\hbar)^3}\int \mathrm{d}\varepsilon\frac{\varepsilon-\mu}{T}\,\frac{\partial f_0(\varepsilon)}{\partial\varepsilon}\int \mathrm{d}p_H \left\{\frac{\partial S}{\partial p_H}\right\}^2 \frac{\cos^2\theta}{\partial S/\partial\varepsilon} \\ &\times\left\{\tau_{\mathrm{osc}} + 2Re\sum_{k=1}^{\infty}(-1)^k\tau_0\int \mathrm{d}p_H\,\left\{\frac{\partial S}{\partial p_H}\right\}^2 \frac{\cos^2\theta}{\partial S/\partial\varepsilon}\exp\left[\frac{\mathrm{i}kcS(\varepsilon,p_H)}{eH\hbar}\right]\right\} \end{aligned} \tag{12}$$

is determined mainly by the oscillatory dependence of τ_{osc}. The terms with $k \neq 0$ in the formula (12) are less than the first term by a factor $\hbar\Omega/\mu$.

After straightforward calculations we obtain the following asymptotic expression:

$$\alpha_{zz}^{\mathrm{mon}} = -\frac{eT\tau_0}{6\hbar^3}\,\frac{\partial}{\partial\mu}\int_{\varepsilon=\mu} m^*\bar{v}_z^2\mathrm{d}p_H, \tag{13}$$

where

$$\begin{aligned} \alpha_{zz}^{\mathrm{osc}} &= \frac{\pi\tau_0 eT}{3\hbar^3}\left(\frac{1}{\hbar\Omega\mu}\right)^{1/2}\sum_{\mathrm{e}}\left|\frac{\partial^2 S_{\mathrm{e}}}{\partial p_H^2}\right|^{-1/2}\sum_{k=1}^{\infty}(-1)^k a_k k^{3/2}\frac{ku}{\sinh(ku)} \\ &\times\cos\frac{\pi k m^*}{m}\,\sin\left(\frac{kcS_{\mathrm{e}}}{e\hbar H} + \frac{\pi}{4}s\right)\int \bar{v}_z^2\mathrm{d}p_H\,. \end{aligned} \tag{14}$$

As is easily seen the amplitude of $\alpha_{zz}^{\rm osc}$ is greater than $\alpha_{zz}^{\rm mon}$ by a factor of $(\mu/\eta\hbar\Omega)^{1/2}$.

The Dingle factor $I_D = \exp(-1/\Omega\tau)$ is omitted in the above formulas because at $(\Omega\tau) \gg 1$ this factor is close to unity. However, as the angle ϑ increases, the period of electron rotation in the magnetic field grows and D decreases. At ϑ close to $\pm\pi/2$ the amplitude of quantum oscillations decays exponentially.

If the magnetic field noticeably deviates from the normal to the layers, both $\alpha_{zz}^{\rm mon}$ and $\alpha_{zz}^{\rm osc}$ oscillate with the angle ϑ between the magnetic field direction and the normal. These oscillations result from the periodic dependence of $\overline{v}_z$ upon the angle ϑ and are characteristic of the kinetic coefficients of layered conductors [9–12].

Using classical equations of motion for electron whose dispersion law is described by the formula (1), in the main approximation in the small parameter η one can easily obtain

$$\overline{v}_z = -\sum_{k=1} \frac{ka}{\hbar}\,\varepsilon_k J_0\left(\frac{kap_{\rm F}\tan\vartheta}{\hbar}\right)\sin\left(\frac{kap_H}{\hbar\cos\vartheta}\right), \tag{15}$$

where J_n is the Bessel function and the coefficients ε_k are assumed to be independent of p_x, p_y. Considering, for example, the monotonously varying part of the field

$$E_z = -\frac{\pi^2 T}{3e}\,\frac{\partial\sigma_{zz}/\partial\mu}{\sigma_{zz}}\,\frac{\partial T}{\partial z}\,, \tag{16}$$

where

$$\sigma_{zz} = \frac{e^2\tau a m^*\cos\vartheta}{2\pi\hbar^2}\sum_{k=1}^{\infty} k^2 J_0^2\left(\frac{akp_{\rm F}\tan\vartheta}{\hbar}\right), \tag{17}$$

we obtain

$$E_z = \frac{\pi^2 T\tan\vartheta}{3e\hbar v_{\rm F}}\,\frac{\sum_{k=1} k^3\varepsilon_k^2 J_0(k\beta)J_1(k\beta)}{\sum_{k=1} k^2\varepsilon_k^2 J_0^2(k\beta)}\,. \tag{18}$$

Here $\beta = (ap_{\rm F}\tan\vartheta)/\hbar$. The functions J_n do not vanish simultaneously, and the more rapid the series in the formula (17)converges the sharper the maxima on the curve $E_z(\vartheta)$ are. The analysis of the angle dependence of the thermoelectric field in layered conductors makes it possible to determine the degree of decreasing of harmonics in the expansion in the Fourier series of the electron energy dependence on the momentum projection on the normal to the layers. It is easy to make sure that the extremum of the thermoelectric field, as a function of $\tan\vartheta$, does not coincide with the maximum of the magnetoresistance. The shift of these extreme values contains an important information about the electron energy spectrum of a layered conductor.

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Термомагнітні явища у шаруватих провідниках

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Теоретично вивчено термоелектричні та термомагнітні явища у шаруватих провідниках з квазідвовимірним електронним енергетичним спектром довільного виду в квантуючому магнітному полі при низьких температурах. Передбачено існування гігантських осциляцій термоелектричного поля як функції оберненої величини магнітного поля, що полегшує експериментальне вивчення квантових осциляційних явищ. Показано, що термоелектричні коефіцієнти шаруватого провідника є періодично залежними від кута між напрямком магнітного поля та нормаллю до шарів. Цей орієнтаційний ефект пов'язаний з квазідвовимірністю енергетичного спектру носіїв заряду і є характерним для шаруватих провідників.

Ключові слова: *термоелектричне поле, шаруватий провідник, квантові осциляції*

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