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NANOSTRUCTURES AND LOW-DIMENSIONAL SYSTEMS

Thermoelectric Effects in Organic Conductors in a Strong Magnetic Field

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Abstract—The linear response of the electron system of a layered conductor to the temperature gradient in this system in a strong magnetic field is investigated theoretically. Thermoelectric emf is studied as a function of the magnitude and orientation of a strong external magnetic field; the experimental investigation of this function, combined with the study of the electric and thermal resistance, allows one to completely determine the structure of the energy spectrum of charge carriers.

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A considerable amount of layered conductors have strongly anisotropic metallic conductivity. The electric conductivity in the plane of layers is several orders of magnitude greater than that along the normal **n** to the layers; this fact is attributed to the quasi-two-dimensional character of the electron energy spectrum. The energy $\varepsilon(\mathbf{p})$ of conduction electrons weakly depends on the projection of their momentum $p_z = \mathbf{n} \cdot \mathbf{p}$ onto the normal **n** to the layers. The Fermi surface (FS) $\varepsilon(\mathbf{p}) = \varepsilon_F$ of a layered conductor is weakly corrugated along the axis p_z and, as a rule, is multisheeted and consists of topologically different elements [1, 2]. According to the energy-band calculations, $(BEDT–TTF)_2Cu(SCN)_2$ and $(BEDT–TTF)_2MHg(SCN)_4$ organic conductors, where M is either a metal from the groups K, Rb, or Tl, or NH_4, have two groups of charge carriers with quasi-two-dimensional and quasi-one-dimensional energy spectra [3]. To understand the electron processes in low-dimensional conductors, one needs detailed information on the energy spectrum of conduction electrons. Electron phenomena in degenerate conductors placed in a strong magnetic field **B** when the rotation frequency of electrons ω_c is much greater than their collision rate $1/\tau$ are very sensitive to the form of the energy spectrum of charge carriers. The investigations of galvanomagnetic phenomena in many layered conductors at low temperatures, when the condition $\omega_c\tau \gg 1$ is satisfied in practically accessible magnetic fields, have allowed one to determine the topological structure of the FS and some details of the electron spectrum in layered structures [1, 2]. Similar information on charge carriers can be obtained by investigating the thermoelectric resistance in a strong magnetic field. The dependence of the kinetic coefficient that relates the thermal flux density to the temperature gradient on the magnitude and orientation of a strong magnetic field does not contain any new information on the spectrum compared with that obtainable from the measurement of electric resistance; however, the investigation of thermoelectric phenomena in a strong magnetic field allows one to obtain essentially new important information on charge carriers; in particular, it allows one to determine the velocity distribution of charge carriers on the FS [4].

Consider a simplest model of a two-band conductor. We will assume that the FS consists of a corrugated cylinder (a quasi-two-dimensional sheet of FS) and corrugated planes oriented so that a plane tangent to a corrugated plane sheet of the FS is parallel to the coordinate plane p_yp_z. Let us represent the dependence of energy on the momentum of conduction electrons on the quasi-two-dimensional sheet of FS as

$$\varepsilon(\mathbf{p}) = \frac{p_x^2 + p_y^2}{2m} + \sum_{n=1}^{\infty} \varepsilon_n \cos\left(\frac{anp_z}{\hbar}\right). \quad (1)$$

Here, $\hbar$ is the Planck constant; a is the interlayer distance; and ε_n rapidly decays as the number n increases, so that the projection of the velocity of electrons from this group onto the normal to the layers,

$$v_z = \frac{\partial\varepsilon}{\partial p_z} = -\sum_{n=1}^{\infty} \varepsilon_n \frac{an}{\hbar} \sin\left(\frac{anp_z}{\hbar}\right) \leq \eta v_F \ll v_F \quad (2)$$

is much less than the maximal velocity in the plane of layers, v_F.

The dependence of the kinetic coefficients on the magnitude of the magnetic field is weakly sensitive to a

specific form of the first term in formula (1); we have chosen this form only to facilitate computations. The electric current induced in the conductor by an external perturbation in the form of an electric field $\mathbf{E}$ and a temperature gradient ∇T,

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

can be determined, for example, by solving the following kinetic equation for the electron distribution function:

$$f(\mathbf{r}, \mathbf{p}) = f_0(\varepsilon) - \left\{\psi_1(\mathbf{r}, \mathbf{p}) + \psi_2(\mathbf{r}, \mathbf{p})\frac{\varepsilon - \mu}{T}\right\}\frac{\partial f_0}{\partial \varepsilon}, \tag{4}$$

where $f_0(\varepsilon)$ and μ are the equilibrium Fermi function and the chemical potential of electrons, respectively; T is temperature in energy units; and the functions ψ_1 and ψ_2 are the solutions of the equations

$$\frac{\partial \psi_1}{\partial t} + \mathbf{v}\frac{\partial \psi_1}{\partial \mathbf{r}} + \hat{W}_p\psi_1 = e\mathbf{E}\cdot\mathbf{v}, \tag{5}$$

$$\frac{\partial \psi_2}{\partial t} + \mathbf{v}\frac{\partial \psi_2}{\partial \mathbf{r}} + \hat{W}_\varepsilon\psi_2 = \mathbf{v}\frac{\varepsilon - \mu}{T}\nabla T. \tag{6}$$

Here, e is the electron charge; the operators $\hat{W}_p$ and $\hat{W}_\varepsilon$ describe the momentum and energy relaxation of electrons, respectively; and t is the motion time of a charge carrier in the magnetic field $\mathbf{B} = (B\cos\varphi\sin\vartheta, B\sin\varphi\sin\vartheta, B\cos\vartheta)$.

In the case of a small-angle scattering of electrons, the momentum relaxation rate of electrons is lower than the energy relaxation rate, and the eigenvalues of the operators $\hat{W}_p$ and $\hat{W}_\varepsilon$ ($1/\tau_p$ and $1/\tau_\varepsilon$, respectively) are essentially different, especially when the charge carriers are scattered mainly by the vibrations of the crystalline lattice. The lower the temperature, the more important is the scattering of electrons by impurity centers in the crystal. As a rule, the effective radius d of the impurity potential forces is on the order of several interatomic distances; i.e., it is less than or on the order of the de Broglie wavelength of electron, $1/k_F$. If the main dissipation mechanism in the system of conduction electrons is the scattering of these electrons by impurity centers, then the times τ_p and τ_ε are of the same order of magnitude, because each collision substantially changes the momentum of an electron. If the potential slowly decays away from the impurity center, i.e., $k_F d \gg 1$, the electrons are scattered at small angles [5], and, just as in the electron–phonon interaction, a single collision is sufficient for the energy relaxation, while the momentum relaxation occurs after a large number of collisions.

We will assume that the magnetic field is sufficiently strong so that the condition $\tau_p \gg \tau_\varepsilon$ holds together with the condition $\omega_c\tau_p \gg 1$ even for $\omega_c\tau_\varepsilon \gg 1$.

The solution of the kinetic equations (5) and (6) for the collision integrals in the τ-approximation allows one to easily determine the kinetic coefficients that relate the electron flows to the electric field and the temperature gradient.

The components of the tensor α_{ij} are related to the components of the electric conductivity tensor σ_{ij} by the simple formula

$$\alpha_{ij} = \frac{\pi^2 T}{3e}\frac{d\tilde{\sigma}_{ij}(\mu)}{d\mu}. \tag{7}$$

In the absence of current ($\mathbf{j} = 0$), the thermoelectric field has the form

$$E_i = \rho_{il}\alpha_{lj}\frac{\partial T}{\partial x_j}, \tag{8}$$

where ρ_{ij} is the electric resistivity tensor, which is the inverse of the electric conductivity tensor σ_{ij}, and the components of $\tilde{\sigma}_{ij}$ coincide with those of σ_{ij} provided that τ_p is replaced by τ_ε in these components.

When there are several groups of charge carriers, one should take into account the contribution of each group to the kinetic coefficients:

$$\sigma_{ij} = \sigma_{ij}^{(1)} + \sigma_{ij}^{(2)}, \quad \alpha_{ij} = \alpha_{ij}^{(1)} + \alpha_{ij}^{(2)}. \tag{9}$$

Here, $\sigma_{ij}^{(1)}$ and $\alpha_{ij}^{(1)}$ are contributions of the charge carriers whose states lie on the plane sheet of FS, and $\sigma_{ij}^{(2)}$ and $\alpha_{ij}^{(2)}$ take into account the contributions of the remaining electrons with the Fermi energy.

The charge carriers whose states belong to the corrugated plane sheet of the FS mainly drift along the x axis and make a significant contribution only to the electric conductivity component σ_{xx}.

The asymptotic value of the component $\sigma_{xx}^{(1)}(B)$ in a strong magnetic field for $\gamma = 1/\omega_c\tau \ll 1$ is of the same order of magnitude as the value of $\sigma_{xx}^{(1)}$ in the absence of the magnetic field:

$$\begin{aligned}\sigma_{xx}^{(1)} \equiv \sigma_1 &= \frac{2e^2}{(2\pi\hbar)^3}\int d\varepsilon dp_y dp_z \frac{v_x^2\tau}{|v_x|}\delta(\varepsilon(\mathbf{p}) - \varepsilon_F) \\ &= \frac{2e^2}{(2\pi\hbar)^3}\int dp_y dp_z |v_x|\tau = \frac{2e^2 v_1\tau}{\pi\hbar ab},\end{aligned} \tag{10}$$

where b is the period of the crystalline lattice along the y axis and v_1 is the modulus of the mean drift velocity of electrons in the x axis. We will assume that v_1 and v_F are of the same order of magnitude.

At sufficiently low temperatures, it is essential that one should take into account the quantization of the energy of charge carriers that perform finite motion in a plane orthogonal to the magnetic field. These are

mainly the charge carriers whose states belong to the quasi-two-dimensional sheet of the FS. If the temperature-induced smearing of the Fermi distribution function is less than the distance between quantized Landau levels, namely, $2\pi^2 T < \hbar\omega_c$, then the contribution of these electrons to the current density

$$\begin{aligned}\mathbf{j} &= \mathrm{Sp}e\hat{\mathbf{v}}\hat{f} \\ &= \frac{2eB}{c(2\pi\hbar)^2}\sum_{n,n'}dp_B e\mathbf{v}_{n,n'}(p_B)f_{n,n'}(p_B)\end{aligned} \tag{11}$$

must be determined either by solving the kinetic equation for the statistic operator $\hat{f}$ [6] or by the Green's function method [7] (see also [8]). In the quasiclassical approximation, when $\hbar\omega_c \ll \eta\mu$, the quantum oscillations of electron flows are largely determined by the oscillation dependence of the scattering amplitude of conduction electrons by impurity centers, i.e., by the quantum oscillations of the mean free path time of the charge carriers [9, 10].

Most conduction electrons whose states belong to the plane sheet of the FS move in a magnetic field along open trajectories; the energy spectrum of these electrons is not discrete–continuous, and their contribution to the kinetic coefficients does not contain quantum corrections.

The components of the tensors $\sigma_{ij}^{(2)}$ and $\alpha_{ij}^{(2)}$ decrease as the magnetic field increases if the value of at least one velocity component v_i or v_j, averaged over the period $2\pi/\omega_c$ vanishes. The charge carriers whose states belong to the quasi-two-dimensional sheet of the FS drift only along the magnetic field provided that the latter significantly deviates from the plane of layers. Thus, when $\gamma_1 = 1/\omega_c\tau_\varepsilon \ll 1$, it suffices to retain only the diagonal matrix elements of the statistical operator $f_{nn}(p_B)$ in the asymptotic expression, say, for α_{zz}; these diagonal elements coincide with the quasiclassical distribution function,

$$\begin{aligned}\alpha_{zz} &= -\frac{2eB}{c(2\pi\hbar)^2} \\ &\times\int dp_B\sum_{n=0}^{\infty} e v_{z,nn}^2(p_B)\tau_\varepsilon\frac{\varepsilon_n-\mu}{T}\frac{\partial f_0(\varepsilon_n)}{\partial\varepsilon_n}.\end{aligned} \tag{12}$$

Applying the Poisson formula

$$\sum_{n=0}^{\infty}\varphi_n = \sum_{k=-\infty}^{\infty}\int dn\varphi(n)\exp(2\pi ikn) \tag{13}$$

and the equation for quasiclassical energy levels

$$S(\varepsilon, p_z) = 2\pi\hbar\frac{eB}{c}\left(n+\frac{1}{2}\right), \tag{14}$$

we obtain the following expressions for $\alpha_{zz} = \alpha_{zz}^{\mathrm{mon}} + \alpha_{zz}^{\mathrm{osc}}$:

$$\alpha_{zz}^{\mathrm{mon}} = \frac{\pi^2 T}{3e}\frac{\partial\tilde{\sigma}_{zz}}{\partial\mu}, \tag{15}$$

$$\begin{aligned}\alpha_{zz}^{\mathrm{osc}} &= -2\mathrm{Re}\sum_{k=1}^{\infty}(-1)^k\frac{2e}{(2\pi\hbar)^3}\int d\varepsilon\frac{\varepsilon-\mu}{T}\frac{\partial f_0}{\partial\varepsilon} \\ &\times\int dp_B 2\pi m^*(\varepsilon, p_B)v_z^2(\varepsilon, p_B)\tau_\varepsilon \\ &\times\exp\left\{\frac{ikcS(\varepsilon, p_B)}{eB\hbar}\right\},\end{aligned} \tag{16}$$

where S is the area of the cross section of the isoenergetic surface by the plane $p_z = \mathrm{const}$ and m^* is the cyclotron effective mass. Only the charge carriers on the corrugated cylinder take part in the formation of quantum oscillations, but both groups of carriers contribute to the slowly varying functions $\sigma_{ij}^{\mathrm{mon}}$ and $\alpha_{ij}^{\mathrm{mon}}$ of the magnetic field.

When $\hbar\omega_c \ll \eta\mu$, we should apply the stationary phase method to carry out integration with respect to p_B in formula (16), as well as in the formula for the oscillating part of the component of the electric conductivity tensor,

$$\begin{aligned}\sigma_{zz}^{\mathrm{osc}} &= -2\mathrm{Re}\sum_{k=1}^{\infty}(-1)^k\frac{2e^2}{(2\pi\hbar)^3}\int d\varepsilon\frac{\partial f_0}{\partial\varepsilon} \\ &\times\int dp_B 2\pi m^*(\varepsilon, p_B)v_z^2(\varepsilon, p_B)\tau_p \\ &\times\exp\left\{\frac{ikcS(\varepsilon, p_B)}{eB\hbar}\right\}.\end{aligned} \tag{17}$$

Then, asymptotically extending the integrand in formula (16) to the domain of complex values of energy and applying the residue theorem, we obtain the final expression

$$\begin{aligned}\alpha_{zz}^{\mathrm{osc}} &= \mathrm{Re}\sum_{k=1}^{\infty}(-1)^k\frac{em^* v_z^2(\mu)\tau_\varepsilon}{\pi^2\hbar^3}\left(\frac{eB\hbar}{kc}\right)^{1/2}\left|\frac{\partial^2 S_e}{\partial p_B^2}\right|^{-1/2} \\ &\times R_D^k Q_T(ku)\exp\left\{\frac{ikcS_e(\mu)}{eB\hbar}+\frac{i\pi}{4}s\right\}.\end{aligned} \tag{18}$$

The asymptotic expression for $\sigma_{zz}^{\mathrm{osc}}$ has the form

$$\begin{aligned}\sigma_{zz}^{\mathrm{osc}} &= \mathrm{Re}\sum_{k=1}^{\infty}(-1)^k\frac{e^2 m^* v_z^2(\mu)\tau_p}{\pi^2\hbar^3}\left(\frac{eB\hbar}{kc}\right)^{1/2}\left|\frac{\partial^2 S_e}{\partial p_B^2}\right|^{-1/2} \\ &\times R_D^k R_T(ku)\exp\left\{\frac{ikcS_e(\mu)}{eB\hbar}+\frac{i\pi}{4}s+\frac{i\pi}{2}\right\}.\end{aligned} \tag{19}$$

Here, S_e is the extremal section of the FS,

$$Q_T = \frac{dR_T(u)}{du}, \quad R_T(u) = \frac{u}{\sinh u},$$
$$u = \frac{2\pi^2 T}{\hbar\omega_c}, \quad s = \operatorname{sgn}\frac{\partial^2 S_e}{\partial p_B^2},$$

and the Dingle factor $R_D^{(k)} = \exp(-k/\omega_c\tau)$ is close to unity and is therefore omitted.

The maxima of the kth harmonics of $\alpha_{ij}^{\mathrm{osc}}(k)$ are shifted by $\pi/2$ relative to the maxima of $\sigma_{ij}^{\mathrm{osc}}(k)$, and the ratio of the amplitudes of these harmonics is

$$\frac{\left|\alpha_{zz}^{\mathrm{osc}}(k)\right|}{\left|\sigma_{zz}^{\mathrm{osc}}(k)\right|} = \frac{Q_T(ku)}{R_T(ku)}\frac{\tau_\varepsilon}{e\tau_p} = \frac{\pi^2 T}{3e\hbar\omega_c}\frac{\tau_\varepsilon}{\tau_p}. \tag{20}$$

The oscillation amplitude of the electric conductivity is $(\eta\mu/\hbar\omega_c)^{1/2}$ times smaller than its slowly varying part, whereas $\alpha_{zz}^{\mathrm{osc}}$ is $(\mu/\eta\hbar\omega_c)^{1/2}$ times greater than $\alpha_{zz}^{\mathrm{mon}}$. A similar relation holds for all the components of $\alpha_{ij}^{\mathrm{osc}}$ and $\alpha_{ij}^{\mathrm{mon}}$ except for the Hall components, which, just as the Hall components of the electric conductivity, do not contain quantum corrections in the collisionless limit $\omega_c\tau_p = \infty$ [11]. As a result, the thermoelectric emf turns out to be an alternating function. This fact allows us to significantly increase the accuracy of determining the extremal sections of the FS by measuring the distance between the maxima (or minima) on the graph of the thermoelectric emf versus $1/B$. The comparison of $\alpha_{ij}^{\mathrm{osc}}$ and $\sigma_{ij}^{\mathrm{osc}}$ allows us to determine the cyclotron effective mass $m^* = (1/2\pi)(\partial S_e/\partial\mu)$ of charge carriers that are responsible for the quantum oscillation effect.

Usually, m^* is determined by measuring the temperature dependence of the amplitude of Shubnikov–de Haas oscillations. To this end, it is necessary that the damping interval of quantum oscillations be neither too small nor large so that the electron–phonon dissipation mechanism of the electron system could not affect the Dingle factor. The determination of the cyclotron effective mass by the joint investigation of magnetoresistance and thermoelectric emf is convenient because there is no need to carry out measurements at different temperatures and it suffices to compare the quantum oscillations of thermoelectric emf with the Shubnikov–de Haas oscillations at constant temperature.

In the case of heating of charge carries along the normal to the layers of a conductor with two-zone spectrum, just as in the case when there is only one group of charge carriers, the thermoelectric field is directed mainly along the temperature gradient,

$$E_z = \left[\frac{\pi^2 T}{3e}\frac{1}{\sigma_{zz}^{\mathrm{mon}}}\frac{\partial\tilde{\sigma}_{zz}^{\mathrm{mon}}}{\partial\mu} + \frac{\alpha_{zz}^{\mathrm{osc}}}{\sigma_{zz}^{\mathrm{mon}}}\right]\frac{\partial T}{\partial z}. \tag{21}$$

The most convenient way to carry out the experimental investigation of thermoelectric phenomena is a configuration when the temperature gradient lies in the plane of layers. In this case, the presence of the FS sheet in the form of a corrugated plane essentially affects the magnitude and orientation of the thermoelectric field. In the main approximation in the small quasi-two-dimensionality parameter η, the thermoelectric field

$$E_y = (\rho_{yx}\alpha_{xj} + \rho_{yy}\alpha_{yj})\frac{\partial T}{\partial x_j} \tag{22}$$

is directed along the y axis and linearly increases with the magnetic field when the temperature gradient is directed along the x axis.

In this case, $\rho_{yy} \approx \sigma_{xx}^{(1)}/(\sigma_{xy}^2 + \sigma_{xx}^{(1)}\sigma_{yy})$ quadratically increases with the magnetic field, whereas α is proportional to $1/B$.

If the temperature gradient is directed along the y axis, then the components E_y and E_z of the thermoelectric field reach saturation in a strong magnetic field, while E_x decreases proportionally to γ:

$$E_x = \frac{\pi^2 T}{3e}\rho_0\gamma\sigma_0\frac{1}{\tau}\frac{\partial\tau}{\partial\mu}\frac{\partial T}{\partial y}, \tag{23}$$

$$E_y = \frac{\pi^2 T}{3e}\rho_0(-\sigma_1 + \gamma^2\sigma_0)\frac{1}{\tau}\frac{\partial\tau}{\partial\mu}\frac{\partial T}{\partial y}, \tag{24}$$

$$E_z = \frac{\pi^2 T}{3e}\tan\vartheta\left[(\sin\varphi - \gamma\cos\varphi)\frac{1}{\sigma_{zz}}\frac{\partial\sigma_{zz}}{\partial\mu} + (\rho_0\sigma_1\sin\varphi + \gamma\cos\varphi)\frac{1}{\tau}\frac{\partial\tau}{\partial\mu}\right]\frac{\partial T}{\partial y}. \tag{25}$$

Here, $\sigma_0 = Ne^2\tau/m$, N is the density of charge carriers on the quasi-two-dimensional sheet of the FS, and $\rho_0 = 1/(\sigma_0 + \sigma_1)$.

In the absence of a plane sheet of the FS, when $\gamma \ll 1$, the thermoelectric field is orthogonal to ∇T and is directed along the normal to the layers, and the components E_x and E_y decrease as the magnetic field increases.

For any orientation of the temperature gradient, the oscillation amplitude of the thermoelectric field is always greater than the part of the thermoelectric emf that slowly varies with the field.

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