

Galvanomagnetic phenomena in layered organic conductors (Review)

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The experimental research on galvanomagnetic phenomena in layered organic conductors at high magnetic fields is discussed in terms of the theoretical ideas about charge transfer phenomena in conductors with a metallic type of conductivity and a quasi-two-dimensional electron energy spectrum of arbitrary form. Attention is devoted mainly to the problem of recovering the dispersion relation of the conduction electrons in layered organic charge-transfer complexes from experimental studies of their magnetoresistance and quantum oscillation phenomena at low temperatures. © 2005 American Institute of Physics. [DOI: 10.1063/1.1884422]

1. INTRODUCTION

Interest in low-dimensional organic conductors rose sharply in the 1960s after Little's suggestion¹ that high-temperature superconductivity might be realized in one-dimensional polymer chains. Despite the fact that this idea has not found experimental confirmation, joint efforts of physicists and chemists have led to the creation of a new class of organic salts having metallic electrical conductivity.^{2,3}

A characteristic feature of the electronic properties of the first organic metals was a pronounced anisotropy of a quasi-one-dimensional type due to their crystal structure. The main structural elements of these compounds are planar molecules having donor or acceptor properties. The best-known examples of such molecules are tetrathiafulvalene (TTF), tetramethyltetraselenafulvalene (TMTSF), tetraselenatetracene (TST), and tetracyanoquinodimethane (TCNQ), which are shown schematically in Fig. 1. The radical ions of these molecules form regular stacks along a preferred crystallographic direction. The interplane distance between molecules is often shortened as compared to the van der Waals separation. The mutual orientation of neighboring radical ions in the stack makes for significant overlap of the π molecular orbitals at minimal Coulomb repulsion. Fractional charge transfer from the radical ions to the counterions causes a partial occupation of the conduction bands thus formed. As a result, the conductivity σ along the stacks at room temperature in a number of compounds exceeds $\sim 10^3$ S/cm and grows with decreasing temperature. At the same time, the overlap of the molecular orbitals between stacks is much weaker, making for extremely low conductivity in the transverse direction, ~ 1 S/cm or less, at room temperature. Such high anisotropy of the electronic properties leads to Peierls instability of the metallic state, characteristic for quasi-one-dimensional conductors: as the temperature is lowered, the substance undergoes a transition to an insulating state with the formation of a charge- or spin-density wave.^{4–6}

To suppress the insulator transition it is necessary to increase the dimensionality of the conducting system, i.e., to strengthen the coupling between stacks. On the one hand, this can be achieved by the application of high pressure. Indeed, a metallic state stable to the very lowest temperatures was first obtained at a pressure $P \approx 5$ kbar in the quasi-one-dimensional complex $(\text{TST})_2\text{Cl}$.⁷ Subsequently similar results were obtained on a number of other compounds.⁸ The most exciting achievement was the discovery in 1980 of superconductivity under pressure ($P \sim 9$ kbar) in $(\text{TMTSF})_2\text{PF}_6$ (Ref. 9) and then in the isostructural salts $(\text{TMTSF})_2\text{X}$ with $\text{X} = \text{AsF}_6, \text{SbF}_6, \text{ClO}_4$, etc. (a detailed

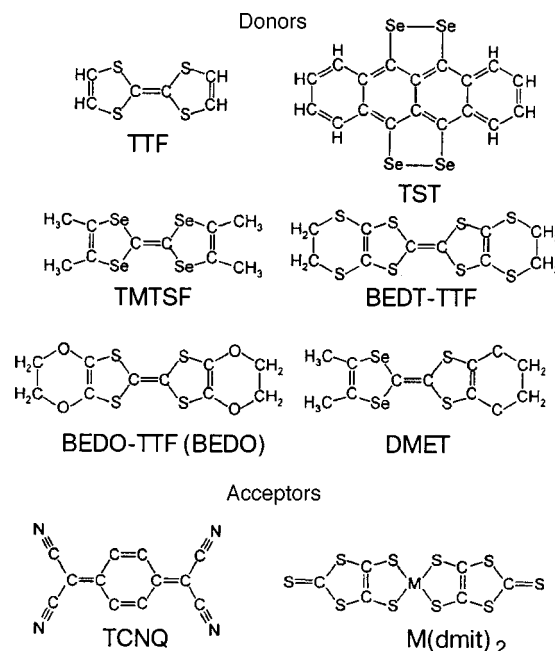


FIG. 1. Donor and acceptor molecules on which the best-known crystalline organic conductors are based (the full names of the molecules are given in the text).

review of the physical properties of these so-called Bechgaard salts can be found in Refs. 8 and 10). It must be noted, however, that the superconductivity in those compounds competes with instability of the Peierls type (in this case leading to a state with a spin-density wave), which limits the temperature of the superconducting transition to values in the 1 K region.

On the other hand, it has proved possible to synthesize conducting complexes in which the organic molecules do not form weakly coupled individual stacks but rather form integral layers with significant overlap of the π orbitals in two directions. For example, the first layered (quasi-two-dimensional) organic superconductor β -(BEDT-TTF)₂I₃ (BEDT-TTF denotes bis(ethylenedithio)tetrathiafulvalene; see Fig. 1) was synthesized in 1984.¹¹ The crystal structure of this compound is given in Fig. 2. The BEDT-TTF^{0.5+} cation radicals form stacks arranged in layers alternating with the layers of I₃⁻ ions. The presence of a significant number of shortened contacts both inside the stacks and between them makes for an almost isotropic conductivity along the layers, $\sigma_{\parallel} \approx 30$ S/cm, at room temperature, while in the direction perpendicular to the layers the conductivity is almost three orders of magnitude lower.¹² Nevertheless the temperature dependence of the resistivity $\rho = 1/\sigma$ is of a metallic character independent of the direction of the current; the resistance falls off monotonically with cooling, and at $T = 2$ K it is more than two orders of magnitude lower than at room temperature. On further cooling the substance goes into

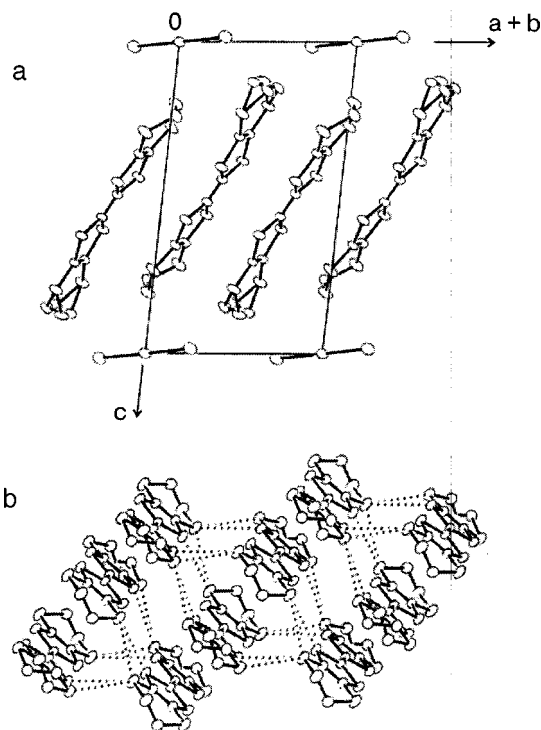


FIG. 2. Crystal structure of the quasi-two-dimensional organic superconductor β -(BEDT-TTF)₂I₃ according to the data of V. F. Kaminskiĭ, T. G. Prokhorova, R. P. Shibaeva, and É. B. Yagubskii, JETP Lett. **39**, 17 (1984). The stacks of BEDT-TTF^{0.5+} cation radicals, which lie in the crystallographic direction ($\mathbf{a} + \mathbf{b}$), form layers separated along the \mathbf{c} axis by layers of I₃⁻ anions (a); the arrangement of the molecules in the conducting layer; the dotted lines denote the shortened contacts responsible for the metallic conductivity between the sulfur atoms from neighboring molecules (b). The figure was kindly provided by S. S. Khasanov and R. P. Shibaeva.

the superconducting state ($T_c \approx 1.5$ K).¹¹ Interestingly, at a relatively low pressure (below 1 kbar) the superconducting transition temperature increases abruptly by a factor of five, jumping from 1.5 K to 7.5–8.0 K.^{13,14}

Soon after the discovery of superconductivity in β -(BEDT-TTF)₂I₃ the isostructural superconducting salts of BEDT-TTF with the anions IBr₂⁻ and AuI₂⁻ were synthesized, with transition temperatures $T_c = 2.7$ K and 4.8 K, respectively,^{15,16} significantly exceeding the highest transition temperature in quasi-one-dimensional superconductors. By now there are some dozens of layered organic superconductors known, most of which are BEDT-TTF salts.¹⁰ Some of the other molecules for which superconducting compounds have been synthesized are shown in Fig. 1: bis(ethylenedioxy)tetrathiafulvalene (BEDO-TTF), dimethylethylenedithio-diselenadithiofulvalene (DMET), and metal complexes of bis-4,5-dimercapto-3-dithiol-2-thione (M(dmit)₂). The record values of T_c at present have been obtained in the layered compounds κ -(BEDT-TTF)₂[N(CN)₂]₂X with X = Br ($T_c \approx 11.6$ K, $P = 0$ kbar)¹⁷ and X = Cl ($T_c \approx 12.8$ K, $P = 0.3$ kbar),¹⁸ and β' -(BEDT-TTF)₂ICl₂ ($T_c \approx 14.2$ K, $P = 82$ kbar).¹⁹

To understand the nature of superconductivity and a number of other, no less interesting, phenomena observed in organic conductors (see the review in Ref. 10, for example), detailed knowledge of the electron band structure of these compounds is needed. In the case of ordinary metals high magnetic fields are a powerful tool for investigating the electronic spectrum. In particular, measurements of the anisotropy of the magnetoresistance permit one to investigate the topology of the Fermi surface of the metal,^{20–24} and from the Shubnikov–de Haas oscillations one can determine the values of the extremal closed cross sections of the Fermi surface and some other important characteristics of the charge carriers.^{23–25} These methods are widely used to study the electronic structure of ordinary three-dimensional metals.^{25–27}

In 1988 Shubnikov-de Haas oscillations were observed in the layered superconductors β -(BEDT-TTF)₂IBr₂ (Refs. 28 and 29) and κ -(BEDT-TTF)₂Cu(NCS)₂ (Ref. 30) at magnetic fields ~ 10 T. These studies provided the first direct proof of the validity of the Fermi liquid description of the electronic properties of the given materials and stimulated intensive further research on organic conductors at high magnetic fields. By the mid-1990s extensive information had been accumulated on the subject, a detailed review of which was given by Wosnitza.³¹ Some interesting results of the application of high fields for studying layered organic conductors are reviewed in Refs. 32 and 33.

By virtue of the extremely high anisotropy of the electronic properties of organic conductors, their behavior in a strong magnetic field differs substantially from that of ordinary three-dimensional materials. This is true of both the quantum oscillations of the magnetoresistance and its quasi-classical components, which demonstrate qualitatively new effects absent in moderately anisotropic three-dimensional metals and in purely two-dimensional conducting systems.

In this article we review the basic galvanomagnetic phenomena observed in layered organic conductors and their use

for quantitative study of the electronic spectrum of these materials. Attention is devoted mainly to the interlayer magnetoresistance features caused by the presence of a Fermi surface in the form of a cylinder with arbitrary cross section and with only a slight corrugation in the direction perpendicular to the highly conductive layers. A prominent example of a system having such a Fermi surface is the superconductor β -(BEDT-TTF)₂IBr₂ (Refs. 10, 31, and 32), which is isostructural to the complex β -(BEDT-TTF)₂I₃. In particular, the Shubnikov-de Haas oscillations with two close frequencies attest to a simply connected Fermi surface in the form of a slightly ($\sim 1\%$) corrugated cylinder occupying approximately half the volume of the Brillouin zone. Such simple topology of the Fermi surface and the high quality of single crystals of this compound make it an excellent model object for studying electronic phenomena in quasi-two-dimensional metals. Below we shall consider the quasiclassical magnetoresistance of β -(BEDT-TTF)₂IBr₂ and show that the effects observed, in particular, the peculiar dependence on the magnetic field direction, are due to the quasi-two-dimensional character of the electron spectrum and permit a quantitative description of the Fermi surface. Section 3 is devoted to Shubnikov-de Haas oscillations, the behavior of which in the substances considered is significantly different from the predictions of the standard three-dimensional theory based on the Lifshitz-Kosevich model. Although a quantitative description of the Shubnikov-de Haas effect for quasi-two-dimensional systems is far from complete, the existing models are capable of explaining a number of qualitative features observed experimentally. In the final Section we give a brief description of the Hall effect expected in a quasi-two-dimensional metal at high magnetic fields.

2. QUASICLASSICAL MAGNETORESISTANCE

The sharp anisotropy of the electrical conductivity of layered conductors is due to anisotropy of the velocities of the conduction electrons $\mathbf{v} = \partial\varepsilon/\partial\mathbf{p}$, and the energy of the charge carriers in such conductors,

$$\varepsilon(\mathbf{p}) = \sum_{n=0}^{\infty} \varepsilon_n(p_x, p_y) \cos\left\{\frac{anp_z}{\hbar} + \alpha_n(p_x, p_y)\right\}; \quad (1)$$

$$\varepsilon_n(-p_x, -p_y) = \varepsilon_n(p_x, p_y);$$

$$\alpha_n(p_x, p_y) = -\alpha_n(-p_x, -p_y)$$

depends weakly on the momentum projection $p_z = \mathbf{p} \cdot \mathbf{n}$ on the normal \mathbf{n} to the layers (a is the distance between adjacent layers, and \hbar is Planck's constant). It is natural to suppose that the functions $\varepsilon_n(p_x, p_y)$ with $n \geq 1$ are much less than the Fermi energy ε_F and fall off rapidly with increasing n , as occurs in the tight-binding approximation, for example.

The charge carrier velocity v_z along the normal to the layers is much less than the characteristic Fermi velocity v_F of the electrons along the layers, and the quasi-two-dimensionality parameter η of the charge-carrier energy spectrum can be determined as the ratio of the maximum value of v_z on the Fermi surface $\varepsilon(\mathbf{p}) = \varepsilon_F$ to the value v_F , i.e.,

$$v_z = - \sum_{n=1}^{\infty} \frac{an}{\hbar} \varepsilon_n(p_x, p_y) \sin\left\{\frac{anp_z}{\hbar} + \alpha_n(p_x, p_y)\right\} \leq \eta v_F. \quad (2)$$

Here the ratio of the transverse conductivity σ_{\perp} to the conductivity σ_0 along the layers in the absence of magnetic field is equal in order of magnitude to the square of the quasi-two-dimensionality parameter, η^2 .

In a magnetic field the components of the conductivity tensor, which relate the current density to the electric field \mathbf{E} ,

$$j_i = \sigma_{ij} E_j, \quad (3)$$

can be found using the Boltzmann transport equation in the τ approximation for the collision integral. Without any model assumptions about the electron energy spectrum, the quasiclassical expression for σ_{ij} in the case of periodic motion of a charge with period $T_B = 2\pi/\omega_c$ in a magnetic field \mathbf{B} has the form

$$\sigma_{ij} = - \frac{2e^3 B}{c(2\pi\hbar)^3} \int d\varepsilon \frac{\partial f_0(\varepsilon)}{\partial \varepsilon} \int dp_B \int_0^{T_B} dt v_i(t) \times \int_{-\infty}^t dt' v_j(t') \exp((t'-t)/\tau). \quad (4)$$

Here t is the time of motion of a conduction electron in the magnetic field under the influence of the Lorentz force

$$d\mathbf{p}/dt = (e/c)[\mathbf{v} \times \mathbf{B}], \quad (5)$$

e , τ , and $f_0(\varepsilon)$ are the charge, mean free time, and equilibrium Fermi distribution function of the conduction electrons, $\omega_c = eB/(m^*c)$ is the cyclotron frequency of an electron in a magnetic field \mathbf{B} , m^* is its cyclotron effective mass, p_B is the momentum projection in the magnetic field direction, and c is the speed of light.

The Fermi surface of layered conductors is weakly corrugated along the p_z axis; it can be multisheet and consist of topologically different elements in the form of slightly corrugated cylinders and slightly corrugated planes in momentum space. In the absence of marked anisotropy of the conductivity in the plane of the layers the most probable shape of the Fermi surface is that of a slightly corrugated cylinder; at least one sheet of the Fermi surface in such layered conductors is a cylinder with cross section located inside one unit cell of momentum space.

Let us consider galvanomagnetic phenomena in a conductor whose Fermi surface is in the form of just one cylinder which is slightly corrugated along the p_z axis, in a magnetic field $\mathbf{B} = (0, B \sin \theta, B \cos \theta)$. The sections of such a surface by the plane $p_B = p_z \cos \theta + p_y \sin \theta = \text{const}$ at $(\pi/2 - \theta) \gg \eta$ are almost the same for different values of the momentum projection p_B on the magnetic field direction, and the velocity components of the conduction electrons in the plane of the layers, $v_x(p_B, t)$ and $v_y(p_B, t)$, depend weakly on p_B . At the same time, the velocity along the normal to the layers is substantially different on different sections of the Fermi surface by the plane $p_B = \text{const}$. Hence it follows that the expansion of the components of the conductivity tensor (4) in power series in the quasi-two-dimensionality parameter η starts with the second or higher power terms, provided that at least one of the indices of σ_{ij} is z (Refs. 34 and 35).

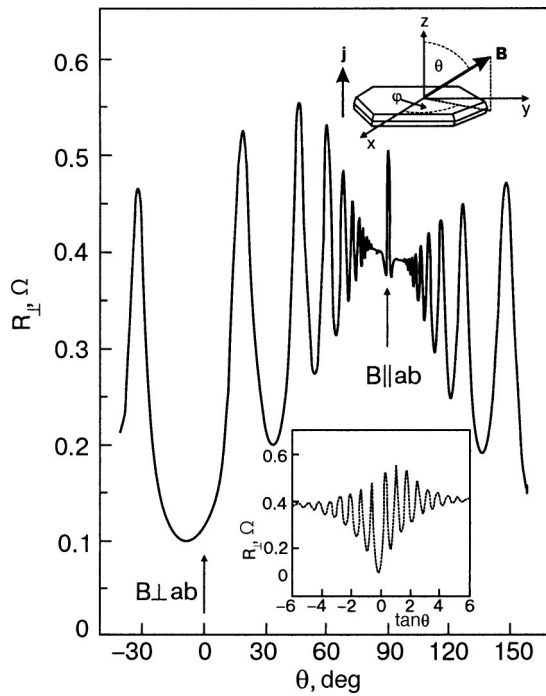


FIG. 3. Resistance of a β -(BEDT-TTF) $_2$ IBr $_2$ single crystal measured in the direction perpendicular to the highly conductive plane ab , in a magnetic field $\mathbf{B}=15$ T, at $T=1.4$ K, versus the angle θ between the field direction and the normal to the ab plane. The geometry of the experiment is illustrated schematically in the upper inset. A characteristic feature of this dependence is the presence of strong oscillations that repeat periodically in the $\tan \theta$ scale, as is shown in the lower inset. In addition, a sharp peak of the magnetoresistance is observed in a narrow neighborhood of angles around $\theta=90^\circ$ (see Sec. 2.2 and Fig. 7).

The resistivity of such conductors along the layers is of the same order of magnitude as that of an uncompensated metal, i.e., at any orientation of the magnetic field the resistivity is essentially no different from that in the absence of field. In contrast, the resistivity ρ_{zz} along the “hard” direction of current flow, i.e., along the normal to the layers, is extremely sensitive to the orientation of a strong magnetic field.

Figure 3 shows an example of the angular dependence of the resistance R_\perp measured in the direction perpendicular to the highly conductive plane ab of a β -(BEDT-TTF) $_2$ IBr $_2$ single crystal as a magnetic field $\mathbf{B}=15$ T is rotated in a plane normal to the ab plane. The geometry of the experiment is shown schematically in the inset. The most remarkable feature of this dependence is obviously the strong oscillations of the magnetoresistance. The positions of the local maxima on the $R_\perp(\theta)$ curve are independent of the magnetic field strength and temperature,³⁶ and, as is seen in the figure, periodically repeat in the $\tan \theta$ scale over the entire range of angles except in a small neighborhood of $\theta=\pi/2$. The field dependence of the magnetoresistance varies sharply as the field direction is changed:^{36,37} as is shown in Fig. 4, the resistance increases in approximate proportion to B^2 for the field direction corresponding to the maximum on the angular dependence (curve 1), while at the minimum a tendency toward saturation in fields above 5 T is clearly seen.

Such behavior, which was first observed²⁹ in a high-quality sample of β -(BEDT-TTF) $_2$ IBr $_2$, turns out to be a general property of quasi-two-dimensional metals and is

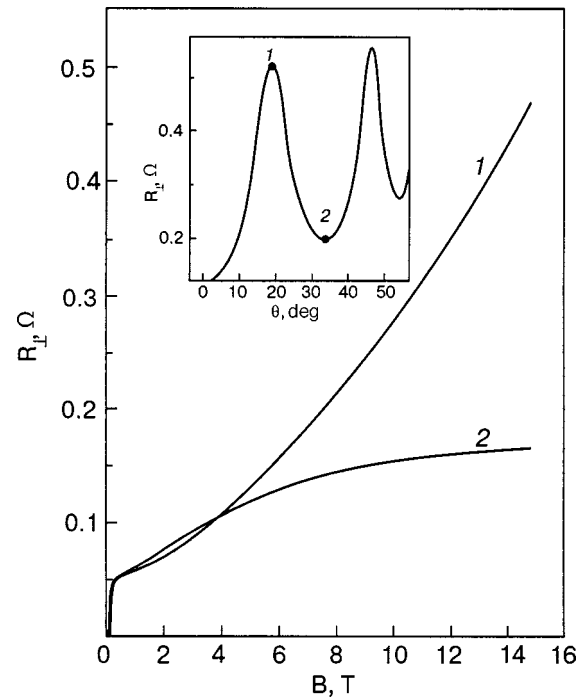


FIG. 4. Field dependence of the interlayer resistance R_\perp of a β -(BEDT-TTF) $_2$ IBr $_2$ single crystal at $T=1.4$ K for two different magnetic field directions. Curve 1 corresponds to a maximum and curve 2 to a minimum on the oscillatory angular dependence, as is shown in the inset.

manifested to some degree or other in practically all layered organic conductors (see, e.g., the review articles^{31–33}) and in a number of other layered structures.^{38–44} This orientation effect does not take place in ordinary metals and is observed only in layered conductors with a quasi-two-dimensional electron energy spectrum.

2.1. Angular oscillations of the magnetoresistance

When current is passed along the normal to the layers the electric field is almost parallel to the current, and ρ_{zz} is equal to $1/\sigma_{zz}$ to within corrections small in the parameter $\eta \ll 1$.

A significant first step in the explanation of the orientation effect was made by Yamaji,⁴⁵ who, for the case of a rather simple charge-carrier dispersion relation

$$\varepsilon = \frac{p_x^2 + p_y^2}{2m} - 2t_\perp \cos\left(\frac{ap_z}{\hbar}\right) \quad (6)$$

calculated, in the linear approximation in the small parameter $\eta \ll 1$, the dependence of the area of section of the isoenergy surface $S(\varepsilon, p_B)$ by a plane $p_B = \text{const}$ on the angle θ between the magnetic field vector and the normal to the layers:

$$S(\varepsilon_F, p_B) \cos \theta = \pi p_F^2 + 4\pi m t_\perp \cos\left(\frac{ap_B}{\hbar \cos \theta}\right) J_0\left(\frac{ap_F}{\hbar} \tan \theta\right), \quad (7)$$

where $p_F = (2m\varepsilon_F)^{1/2}$, and $J_0(u)$ is the zeroth-order Bessel function. It is clear from expression (7) that the areas of all the cyclotron orbits are practically equal at the periodically repeating zeroes of the Bessel function. Obviously this can

be achieved by an appropriate choice of the angle θ_c . At such an angle the drift velocity of the charge carriers,⁴⁶

$$\bar{v}_z(p_B) = T_B^{-1} \int_0^{T_B} dt v_z(t, p_B) = \cos \theta \frac{\partial S / \partial p_B}{\partial S / \partial \varepsilon} \quad (8)$$

becomes negligibly small. This leads to a sharp decrease in the conductivity σ_{zz} and ultimately to a sharp peak of the magnetoresistivity ρ_{zz} at $\theta = \theta_c$.

Formulas (7) and (8) correctly reflect the qualitative nature of the angular oscillations of the magnetoresistance observed experimentally. In fact, the sharp decrease of the difference between the maximum area S_{\max} and minimum area S_{\min} of the cross section of the Fermi surface at the maxima of the angular dependence was evidenced back in the experiment of Ref. 29: at those orientations of the magnetic field for which the magnetoresistance took a maximum value the beats of the Shubnikov-de Haas oscillations, due to the difference ($S_{\max} - S_{\min}$), vanished.

The conductivity tensor component σ_{zz} at high magnetic field ($\omega_c \tau \gg 1$) in the case of the charge-carrier dispersion relation (6) with corrections taking into account small parameters η and $\gamma = 1/\omega_c \tau$ has the form^{34,47}

$$\sigma_{zz} = \frac{2ae^2 m^* \tau \cos \theta}{\pi \hbar^4} I_{\perp}^2 J_0^2 \left(\frac{ap_F}{\hbar} \tan \theta \right) + \eta^2 \sigma_0 (\eta^2 \Phi_1 + \gamma^2 \Phi_2), \quad (9)$$

where σ_0 is the conductivity along the layers in the absence of magnetic field, and Φ_1 and Φ_2 are functions of the tilt angle of the magnetic field to the layers and are of the order of unity.

For arbitrary field direction the Bessel function J_0 is generally nonzero, and the conductivity σ_{zz} is determined by the first term in expression (9). In that case the magnetoresistance is essentially the same as that observed in ordinary uncompensated metals: it is relatively low and goes to saturation at high magnetic field. For $\theta = \theta_c$, however, when $J_0 = 0$, the conductivity is proportional to γ^2 , and the magnetoresistance to current transverse to the layers grows with magnetic field in proportion to B^2 , reaching saturation only in the region of very high magnetic fields, i.e., for $\gamma \ll \eta$. Such a character of the field dependence is in good qualitative agreement with the experimental results presented in Fig. 4.

Naturally, a theoretical analysis of the transport phenomena with the use of a very simple model of the carrier dispersion relation in the form (6) cannot claim to give a quantitative description of the experimentally observed dependence of the magnetoresistance on the strength of the magnetic field and on its orientation with respect to the crystallographic axes; nevertheless, in many cases this model of the electron energy spectrum permits a correct comprehension of the nature of electronic phenomena in layered conductors.

In the case of an arbitrary dispersion relation of the charge carriers an asymptotic expression for $\sigma_{zz}(\eta, \gamma)$ at arbitrarily small γ and η has the form.^{34,48}

$$\sigma_{zz} = \sum_{n=1}^{\infty} \int_0^{T_B} dt \int_{-\infty}^t dt' \left(\frac{an}{\hbar} \right) \varepsilon_n(t) \varepsilon_n(t') \frac{e^3 B \cos \theta}{ac(2\pi\hbar)^2} \times \exp\left(\frac{t' - t}{\tau} \right) \cos\left[\frac{an}{\hbar} [p_y(t) - p_y(t')] \right], \quad (10)$$

where all the functions in the integrand are functions of t and t' only. With corrections small in the parameters $\gamma \ll 1$ and $\eta \ll 1$ taken into account, the conductivity tensor component σ_{zz} takes the following form.^{48,49}

$$\sigma_{zz} = \frac{ae^2 m^* \tau \cos \theta}{2\pi\hbar^4} \sum n^2 I_n^2(\theta) + \eta^2 \sigma_0 (\eta^2 \varphi_1 + \gamma^2 \varphi_2); \quad (11)$$

where

$$I_n(\theta) = T_B^{-1} \int_0^{T_B} dt \varepsilon_n(t) \cos\{nap_y(t) \tan \theta / \hbar\}, \quad (12)$$

and the functions φ_1 and φ_2 , which depend on the orientation of the magnetic field, are of the order of unity, as in the case of the carrier dispersion relation (6). The main contribution to the integral $I_n(\theta)$ for $\tan \theta \gg 1$ comes from small neighborhoods of points of stationary phase, where $\partial p_y / \partial t = -(eH/c)v_x \cos \theta = 0$. There are at least two such points on the closed electron orbit: these are the turning points where $v_x(t_{1,2}) = 0$. Here $\varepsilon_n(t_1) = \varepsilon_n(t_2)$ and, if there are no other points of stationary phase on the electron orbits, the asymptotic expression for $I_n(\theta)$ takes the form

$$I_n(\theta) = 2\varepsilon_n(t_1) \frac{|2\pi\hbar|^{1/2}}{T_B |anp_y''(t_1) \tan \theta|^{1/2}} \times \cos\left[\frac{nap_y(t_1)}{\hbar} \tan \theta - \frac{\pi}{4} \right], \quad (13)$$

where a prime denotes differentiation with respect to time. The functions $I_n(\theta)$ have a set of zeroes which for $\tan \theta \gg 1$ repeat with a period^{34,48}

$$\Delta(\tan \theta) = 2\pi\hbar / aD_p, \quad (14)$$

where $D_p \equiv 2p_y(t_1)$ is the extent of the cross section of the Fermi surface along the p_y axis. Thus for an arbitrary form of the quasi-two-dimensional electron energy spectrum for $\tan \theta \gg 1$ the conductivity transverse to the layers, expressed by formula (11), and, hence, the interlayer resistance vary with period (14) on increasing $\tan \theta$.

From the periods of the angular oscillations of the magnetoresistance for different orientations of a strong magnetic field one can determine the shape of the cross section of the cylindrical Fermi surface. Such a procedure was first applied to β -(BEDT-TTF)₂IBr₂ in Ref. 46. The result is presented in Fig. 5. This effect is now widely used to study the Fermi surfaces of organic metals and other layered conductors (see, e.g., Refs. 31–33, 43, and 44).

Naturally, all the terms in the sum over n in formula (11) cannot vanish simultaneously. For example, at $\theta = \theta_1$, when $I_1(\theta)$ vanishes, all the functions $I_n(\theta)$ for which $(n-1)$ is not a multiple of four are substantially nonzero,⁴⁹ and the asymptotic behavior of σ_{zz} depends substantially on the character of the decay of the functions $\varepsilon_n(p_x, p_y)$ with in-

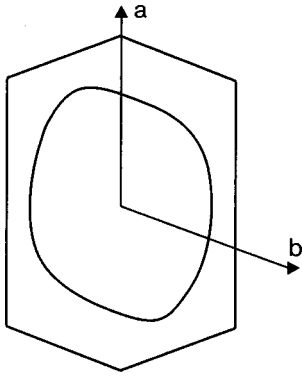


FIG. 5. Cross section of the Fermi surface of the layered organic superconductor β -(BEDT-TTF) $_2$ IBr $_2$, determined from experiment on the angular oscillations of the magnetoresistance.⁴⁶ The Brillouin zone boundary and the directions of the crystallographic axes in the plane of the layers are shown.

creasing index n . Agreement with experiment can be achieved by keeping certain terms in the sum over n in the formula (11) for σ_{zz} .

A theoretical calculation of the resistance to current transverse to the layers with three terms retained in the sum over n in the formula (11) for σ_{zz} gives a result for the θ dependence of the magnetoresistance of the conductor β -(BEDT-TTF) $_2$ IBr $_2$ that is closer to the experimentally observed dependence if $I_{n+1}/I_n=0.4$, while for the more anisotropic organic conductor (BEDT-TTF) $_2$ DIA (DIA is diiodoacetylene) this is found for $I_{n+1}/I_n=0.2$ (Ref. 50). Figure 6 shows the results of a calculation of the angular dependence of the magnetoresistance of the organic conductor β -(BEDT-TTF) $_2$ IBr $_2$ with several harmonics kept in the dispersion relation of the charge carriers for $I_{n+1}/I_n=0.04$ and $I_{n+1}/I_n=0.4$. The experimental angular dependence of the magnetoresistance was taken from Ref. 46.

The energy spectrum of the charge carriers in almost all organic compounds lacks symmetry with respect to the replacement of p_z by $-p_z$, and taking the phase of $\alpha_n(p_x, p_y)$ into account in formula (11) has a sensitive effect on the position of the sharp maxima of the magnetoresistance ver-

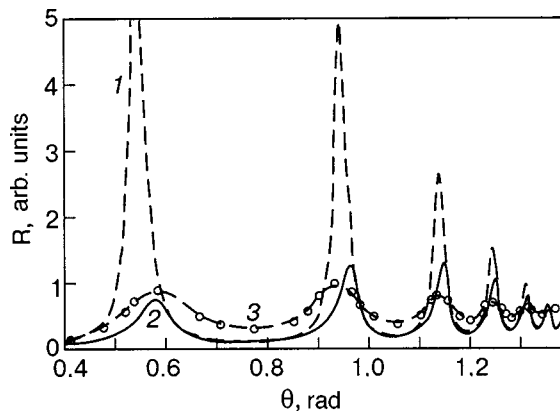


FIG. 6. Dependence of the magnetoresistance transverse to the layers for the organic conductor β -(BEDT-TTF) $_2$ IBr $_2$ on the angle θ between the magnetic field vector and the normal to the layers, calculated theoretically with several Fourier components taken into account in the dispersion relation of the charge carriers: curve 1—for $I_{n+1}/I_n=0.04$; curve 2—for $I_{n+1}/I_n=0.4$. Curve 3—experimental curve of the angular dependence of the resistance transverse to the layers.⁴⁶

sus θ . In Ref. 46, by writing $\alpha_1(p_x, p_y)$ in the form of a linear combination of p_x and p_y , the results of the calculation were reconciled with the experimentally observed asymmetry in the angular dependence of the magnetoresistance.

2.2. Resistance in a magnetic field almost parallel to the layers

The contribution to the conductivity from the rapidly oscillating functions in the integrand in formula (12) for $\gamma \ll 1$ is smaller the larger the value of $\tan \theta$. Consequently, the monotonic part of the magnetoresistance increases with deviation of the magnetic field from the normal to the layers in proportion to $\tan \theta$, as long as $\eta \tan \theta \ll 1$.

When θ is quite close to $\pi/2$, specifically for $\eta \tan \theta \approx 1$, a necking of the electron orbit occurs along the p_x axis. In this region of angles θ a substantial rearrangement of the electron orbits occurs. When the neck width Δp goes to zero, a small orbit splits off from the highly elongated orbit; this small orbit is located completely inside one unit cell of momentum space. The nucleation of small electron orbits begins at a parabolic point of the Fermi surface, where the maximum value of the electron velocity along the magnetic field on the electron orbit occurs. In the case of charge-carrier dispersion relation (6) the small electron orbits arise when $\cos \theta = \eta$, and for an arbitrary shape of the Fermi surface in the form of a weakly corrugated cylinder, when $\cos \theta$ is of the order of η . With further growth of θ the number of small orbits increases, and at $\theta = \pi/2$ the relative fraction of charge carriers with small orbits in momentum space becomes of the order of $\eta^{3/2}$.

The character of the angular dependence of the magnetoresistance for $\eta \tan \theta \gg 1$ is easily ascertained for arbitrary dependence of the energy of the charge carriers on their momentum. In this region of magnetic-field tilt angles with respect to the layers the values of p_x , p_y , and v_x vary slowly with time, while p_z , to a sufficient degree of accuracy (to corrections small in η and $\cos \theta$) varies in time by a linear law almost everywhere on the slightly elongated orbits except in the vicinity of necks and turning points, where the velocity projection v_x is small.

To calculate the conductivity tensor component σ_{zz} for $\eta \tan \theta \approx 1$ we use the Fourier representation for the electron velocity along the z axis:

$$v_z(t) = \sum_{k=-\infty}^{\infty} v_z^{(k)} \exp(ik\omega_c t). \quad (15)$$

The contribution to the conductivity along the normal to the layers:

$$\sigma_{zz} = \frac{2e^2\tau}{(2\pi\hbar)^3} \times \int_0^{2\pi\hbar \cos \theta/a} 2\pi m^* dp_B \left\{ \frac{1}{v_z^2} + 2 \sum_{k=1}^{\infty} \frac{(v_z^{(k)})^2}{1 + (k\omega_c\tau)^2} \right\} \quad (16)$$

from charge carriers executing motion along orbits with small necks is large, since they travel near the neck for a long time. Their period of gyration in a magnetic field diverges logarithmically as Δp goes to zero: $T_B \propto \ln(1/\Delta p)$, so

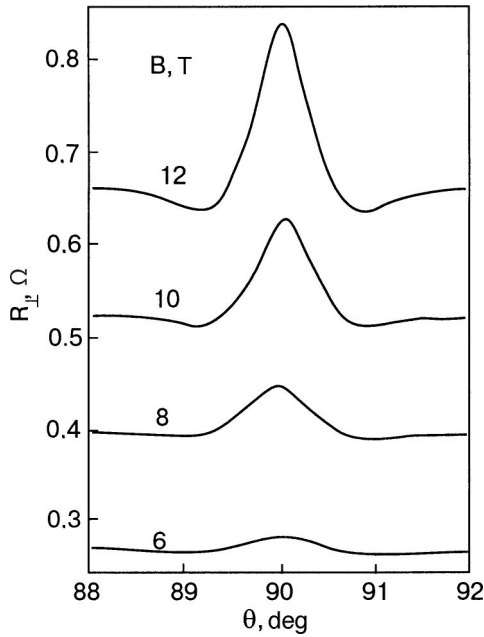


FIG. 7. Structure of the peak on the angular dependence of the magnetoresistance of β -(BEDT-TTF) $_2$ I $_3$ in the vicinity of $\theta=90^\circ$ for different values of the magnetic field. Plotted according to the data of Ref. 53.

that in a certain layer of electron orbits this period is much greater than the mean free time. The lower the cyclotron frequency ω_c , the more terms must be taken into account in the sum over k in formula (16), and for $\omega_c \rightarrow 0$ the contribution to σ_{zz} of such electrons is comparable to their contribution in the absence of magnetic field. Thus the appearance of self-intersecting electron orbits for $\eta \tan \theta \approx 1$ leads to improvement of the conductivity of the layered conductor.^{51,52} As the angle θ approaches closer to $\pi/2$ the magnetoresistance begins to grow, since the term v_z , which is independent of the magnetic field strength, goes to zero in proportion to $\cos^2 \theta$ and the resistance to current transverse to the layers, after passing through a minimum in the region of angles where $\cos \theta$ is of the order of η , again increases, reaching its maximum at $\theta = \pi/2$.^{51,52} In this case the resistance to current transverse to the layers increases without saturation with increasing magnetic field in the plane of the layers.

Formula (16) gives a good description of experiment. Indeed, the angular dependence of the magnetoresistance in Fig. 3 demonstrates a slight drop that is followed by a rapid rise at angles $\theta \rightarrow \pi/2$. A detailed study of the magnetoresistance peak in the vicinity of $\theta = \pi/2$ was carried out by Hanasaki *et al.*⁵³ for the layered conductor β -(BEDT-TTF) $_2$ I $_3$, a close analog of the complex β -(BEDT-TTF) $_2$ IBr $_2$. In particular, it was found experimentally that the width of the peak is practically unchanging with magnetic field strength. This is illustrated in Fig. 7, where the angular dependence of the magnetoresistance β -(BEDT-TTF) $_2$ I $_3$ at various values of the field is shown for angles close to $\pi/2$. The constancy of the peak width permitted the authors of Ref. 53 to attribute its origins to the geometry of the Fermi surface, specifically, to the slight corrugation along the z axis, and thus to estimate the quasi-two-dimensionality parameter $\eta \approx 10^{-2}$.

Let us now consider the dependence of the resistance on the magnetic field strength for $\theta = \pi/2$. The main contribu-

tion to the conductivity tensor component σ_{zz} at $\theta = \pi/2$ is from the small fraction of conduction electrons with orbits near the self-intersecting cross section $p_B = p_c$ of the Fermi surface. These charge carriers move slowly along the z axis with a period $T(p_B)$ larger than or comparable to their mean free time τ for arbitrarily high values of the magnetic field. Since the velocity along the x axis for electrons on orbits $p_B = p_y$ close to p_c is small, which corresponds to a weak dependence of ε on p_x , in calculating the period $T(p_y)$ of the electron's motion one is justified in using an expansion of the energy in a power series for small p_x , dropping the higher-order harmonics in formula (1):

$$\varepsilon = \varepsilon_0(0, p_y) + p_x^2/2m_1 + \varepsilon_1(0, p_y)\cos(ap_z/\hbar). \quad (17)$$

Using relation (17), one can easily calculate the period of the electron's motion along orbits close to self-intersecting,

$$T_B(p_y) = \eta^{-1/2} T_0 \int_0^\pi d\alpha (\xi^2 + \sin^2 \alpha)^{-1/2}, \quad (18)$$

where $T_0 = 2\pi\hbar c/(aeBv_F)$ agrees in order of magnitude with the period of a conduction electron's motion in a magnetic field normal to the layers, and

$$\xi^2 = [\varepsilon - \varepsilon_0(0, p_y) - \varepsilon_1(0, p_y)]/2\varepsilon_1(0, p_y). \quad (19)$$

As the self-intersecting orbit is approached, ξ becomes an arbitrarily small quantity, and the integral in formula (18) diverges logarithmically in proportion to $\ln(1/\xi)$.

Unlike ordinary metals, where the period of the carriers' motion is greater than or comparable to the mean free time only in an exponentially small region of the cross sections of the Fermi surface near the self-intersecting orbit,⁵⁴ in a quasi-two-dimensional conductor the condition $T_B > \tau$ is valid in a significantly wider region of electron orbits, since the period of the electrons' motion near the self-intersecting orbit, even for ξ of the order of unity, is inversely proportional to the small parameter $\eta^{1/2}$. Thus for $\eta^{1/2} \leq \gamma_0 \ll 1$, where $\gamma_0 = T_0/\tau$, there are quite many charge carriers whose period of motion in the magnetic field are greater than or comparable to the mean free time. As a result of averaging over states of the conduction electrons the conductivity of a layered conductor falls off in proportion to $1/B$ with increasing magnetic field directed in or near the plane of the layers.³⁵

$$\sigma_{zz} = \eta^2 \sigma_0 \gamma_0. \quad (20)$$

With increasing magnetic field the number of electrons whose periods of motion exceed the mean free time decreases, but in the limit of high magnetic field, when $\gamma_0 \ll \eta^{1/2} \ll 1$, the contribution of the small fraction of electrons with open trajectories in momentum space close to the self-intersecting cross section of the Fermi surface,

$$\sigma_{zz} = \eta^{3/2} \sigma_0 \gamma_0^2, \quad (21)$$

nevertheless exceeds the contribution to σ_{zz} from all the other electrons. In this region of magnetic fields the linear growth of the resistance to current transverse to the layers gives way to a quadratic growth with magnetic field.^{35,48}

In formulas (20) and (21) we have dropped numerical factors of order unity which depend on the concrete form of the electron energy spectrum.

An analogous dependence of σ_{zz} on magnetic field strength was obtained by Lebed and Bagmet⁵⁵ and by Schofield and Cooper⁵⁶ with the use of a model of the form (6) for the electron energy spectrum.

In the case of a carrier dispersion relation (6) the dependence of the momentum component p_z on time τ in a magnetic field parallel to the plane of the layers is described by the standard equation for a simple pendulum:

$$\frac{\partial^2 p_z}{\partial t^2} + \frac{2at_\perp e^2 B^2}{mc^2 \hbar} \sin \frac{ap_z}{\hbar} = 0. \quad (22)$$

The solution of this equation enables one to write the dependence of the electron velocity v_z on time t explicitly with the aid of the Jacobi functions, which together with their Fourier transforms have been tabulated in sufficient detail, and Schofield and Cooper⁵⁶ had no difficulty in carrying out a numerical calculation of the dependence of the resistance to current transverse to the layers on the magnetic field strength over a wide range of magnetic field and parameter η . It follows from their calculation that in a quasi-two-dimensional conductor ($\eta \ll 1$) the contribution to σ_{zz} from electrons with small orbits for $\theta = \pi/2$ over a wide region of magnetic fields is significantly smaller than the contribution of the charge carriers with open trajectories in momentum space, which thus govern the behavior of the magnetoresistance at high magnetic fields, in accordance with what we have said above.

Concluding this Section, we note that the unrestricted growth of the magnetoresistance with in-plane field is due to the absence of drift of the charge carriers along the current direction, i.e., along the z axis,²⁰ whereas in a magnetic field tilted with respect to the layers the growth of the resistance to current transverse to the layers with increasing magnetic field, $\rho(B) = \rho_{zz} = 1/\sigma_{zz}$, gives way to saturation at high fields:

$$\rho(\infty) = 1/\langle \bar{v}_z^2 \tau \rangle. \quad (23)$$

The angle brackets denote integration over the Fermi surface with weight factor $2e^3 B/c(2\pi\hbar)^3$.

For any magnetic field orientation ρ_{zz} increases with magnetic field, since all the diagonal components of the conductivity tensor fall off monotonically with increasing magnetic field. One can readily see this by turning to formula (16), from which it follows that $\sigma_{zz}(0) \geq \sigma_{zz}(B)$, and

$$\frac{\partial \sigma_{zz}(B)}{\partial B} = - \frac{8e^2 \tau}{(2\pi\hbar)^3 B} \times \int_0^{2\pi\hbar \cos \theta/a} 2\pi m^* dp_B \left\{ \sum_{k=1}^{\infty} \frac{(v_z^k \omega_c \tau)^2}{[1 + (k\omega_c \tau)^2]} \right\} \leq 0, \quad (24)$$

where the equals sign applies only in the case of the longitudinal conductivity of an isotropic conductor.

The rate of increase of the resistivity ρ_{zz} with magnetic field depends substantially on the saturation value (23) to which the resistivity tends in the limit of infinitely high magnetic field. At the maximum of the angular dependence of the resistivity the value of $\langle \bar{v}_z^2 \tau \rangle$ is proportional to η^4 , and the resistivity increases more strongly with magnetic field B than

at the minima of $\rho(\theta)$. At those magnetic-field orientations for which $\rho(\theta)$ has a minimum it is significantly easier to achieve saturation with respect to magnetic field strength, as is indeed observed experimentally^{36,37,57} (see Fig. 4).

3. SHUBNIKOV-DE HAAS EFFECT

With decreasing temperature the mean free path of the charge carriers increases, and the condition of high magnetic field ($\omega_c \tau \gg 1$), under which the dynamic properties of the conduction electrons are most clearly manifested, is realized in a wider range of magnetic fields. At very low temperatures, however, the quasiclassical treatment of the transport properties can turn out to be incorrect.

At Leiden in 1930 Shubnikov and de Haas observed a complicated magnetic-field dependence of the resistance at 20 K in a single-crystal bismuth sample of very high quality for that time.⁵⁸ Against the background of significant growth of the resistivity of bismuth with magnetic field a tendency toward oscillatory behavior of ρ versus B was seen, and an investigation at liquid helium temperature revealed a periodic dependence of magnetoresistance of bismuth on the inverse magnetic field.⁵⁹

This effect, which came to be called the Shubnikov-de Haas effect, did not follow from a quasiclassical treatment of charge transport phenomena in solids and for a long time was regarded as another anomaly among the unusual properties of bismuth. It was only after 8 years that Landau showed⁶⁰ that oscillatory dependence of the magnetoresistance on $1/B$ and also the oscillations of the magnetic susceptibility of bismuth, which had been discovered by de Haas and van Alphen in 1930 as well,⁶¹ are due to quantization of the energy of the charge carriers in a magnetic field and are inherent to all degenerate conductors. In a quantizing magnetic field the density of states of the electrons has a square-root singularity, which at the Fermi level repeats periodically with variation of $1/B$; this is what leads to the oscillatory dependence of the thermodynamic and kinetic characteristics of the conductor on the inverse magnetic field. Rather high magnetic fields are needed for observation of these oscillations, sufficient that the distance between the quantized Landau levels, $\Delta \varepsilon = \hbar \omega_c$, exceeds their width \hbar/τ , and the temperature smearing of the Fermi distribution function $k_B T$ (k_B is Boltzmann's constant) but nevertheless much less than the Fermi energy ε_F , i.e., the condition $k_B T \leq \hbar \omega_c \ll \varepsilon_F$ must be met. In metals the carriers responsible for these quantum oscillations comprise only a small fraction, of the order of $(\hbar \omega_c / \varepsilon_F)^{1/2}$, made up of those for which the area of section $S(\varepsilon, p_B)$ of the Fermi surface $\varepsilon(\mathbf{p}) = \varepsilon_F$ by the plane $p_B = \text{const}$ is close to the extremal value S_{ext} .

From the period of the magnetization or magnetoresistance oscillations,

$$\Delta(1/B) = \frac{2\pi\hbar e}{cS_{\text{ext}}}, \quad (25)$$

one can determine the extremal area of plane sections of the Fermi surface. Thus a reliable spectroscopic method was developed^{62–64} which is still being used successfully to reconstruct from experimental data the main characteristic of

the electron energy spectrum of degenerate conductors—the Fermi surface (see, for example, the monograph by Shoenberg²⁵).

Oscillatory dependence of the magnetoresistance of a metal on $1/B$ due to quantization of the energy of the orbital motion of the charge carriers in a magnetic field was first calculated by Akhiezer⁶⁵ using Titeica’s method.⁶⁶ The essence of Titeica’s method is that by taking into account the oscillatory character of the motion of electrons in a magnetic field, one can represent the electric current as the drift of the centers of the electron orbits. Here the resistance to electrical current flowing in the direction orthogonal to a strong magnetic field arises because of scattering of the charge carriers. Akhiezer, following Titeica, assumed that the mechanism of dissipation in the system of conduction electrons was their scattering by phonons. Although Akhiezer’s work contained a number of errors in the calculations of the amplitude of the magnetoresistance oscillations,⁶⁷ he nevertheless obtained the correct expression for the period of the oscillations and pointed out the significant growth of the quantum corrections to the magnetoresistance as the temperature approaches zero.

The quantum oscillation effects in the magnetoresistance of bismuth at extremely low temperatures, when the charge carriers are scattered mainly by impurity atoms, were considered by Davydov and Pomeranchuk.⁶⁸ Already in that paper it was shown that the probability of scattering of an electron will oscillate with variation of the magnetic field and that it is extremely important to take such oscillations into account. Zil’berman⁶⁹ applied the Titeica’s method in calculating the quantum oscillations of the magnetoresistance in the case of scattering of conduction electrons in a metal by heavy impurities and showed that the amplitude of the Shubnikov-de Haas oscillations for $\varepsilon_F/\hbar\omega_c \gg 1$ is determined mainly by the oscillatory dependence of the mean free time of the charge carriers on $1/B$. The magnetic fields available in the 1950s were not very high, and experimental research on electronic phenomena was restricted to fields of several tesla. Therefore Zil’berman’s rather cumbersome and detailed calculations were confined to the use of formulas for the electron collision frequency and magnetoresistance valid for $k_B T \gg \hbar\omega_c$. The formula he obtained for the inverse mean free time of electrons in a quantizing magnetic field had the form

$$\frac{1}{\tau} = \frac{1}{\tau_0} \left\{ 1 + \frac{9\hbar\omega_c}{40\varepsilon_F} - \frac{5\sqrt{2}}{\sqrt{\varepsilon_F\hbar\omega_c}} \pi^2 k_B T \right. \\ \left. \times \exp\left(-\frac{2\pi^2 k_B T}{\hbar\omega_c}\right) \cos\left(\frac{2\pi\varepsilon_F}{\hbar\omega_c} - \frac{\pi}{4}\right) \right\}. \quad (26)$$

Later Titeica’s method was refined substantially by Kubo⁷⁰ and successfully used in a theoretical study of galvanomagnetic phenomena in metals in a quantizing magnetic field.⁷¹

In the theoretical papers mentioned above^{65–69} the energy spectrum of the conduction electrons was assumed isotropic. Such a model of the carrier dispersion relation — the Drude-Lorentz-Sommerfeld model—was used in those years in many theoretical investigations of electronic phenomena in metals, even though the most probable open Fermi surfaces for electrons in metals with a face-centered cubic lat-

tice had already been proposed in the monograph by Bethe and Sommerfeld (see Figs. 23–25 of Ref. 72). A quadratic isotropic energy spectrum of the charge carriers was also used later in the paper by Adams and Holstein,⁷³ which was devoted to the study of galvanomagnetic phenomena in conducting media.

In the case of an isotropic dispersion relation of the charge carriers there is only one extremal plane section of the Fermi surface—the central cross section, of area $S_{\max} = \pi p_F^2 = 2\pi\varepsilon_F m^*$.

Low-temperature experiments on the magnetic susceptibility of rather pure metals, carried out by Shoenberg and co-workers at Cambridge and by Verkin and Lazarev and co-workers at Kharkov (see Ref. 25 and the references cited therein), and also resonance and magnetoacoustic phenomena^{23,24} have given evidence that even in ordinary quasi-isotropic metals, except for a small group of alkali metals, the electron energy spectrum is rather complex and is substantially different from the spectrum of free electrons.

To explain the experimental studies of quantum oscillation phenomena it was necessary to create a theory with the real electron energy spectrum taken into account. The success of Lifshitz and Kosevich’s theory⁶⁴ of the de Haas-van Alphen effect under the most general assumptions about the form of the electron energy spectrum of metals with the use of only the area quantization rule

$$S(\varepsilon, p_B) = 2\pi\hbar(n + 1/2)eB/c, \quad (27)$$

where n is a nonnegative integer, stimulated investigation of the electronic properties of metals without the invocation of model assumptions about the charge carrier dispersion relation.

One of the first papers devoted to the theoretical study of galvanomagnetic effects in a quantizing magnetic field in conductors with an arbitrary carrier dispersion relation was that of Lifshitz.⁷⁴ In it the current density

$$\mathbf{j} = \text{Tr}(e\hat{\mathbf{v}}\hat{f}) \quad (28)$$

was found by solution of the quantum kinetic equation for the single-particle statistical operator or density matrix \hat{f} , linearized with respect to a weak perturbation of the electronic system by a uniform external electric field. The theory of quantum phenomena in metals with an arbitrary carrier dispersion relation obtained its further development in the paper by Kosevich and Andreev,⁷⁵ who calculated in the Born approximation the correction oscillatory in $1/B$ to the collision integral in the case of electron scattering by impurity atoms with a short-range potential with the use of the Bogolyubov method.⁷⁶ Here the oscillatory (in $1/B$) dependence of the eigenvalues of the collision operator differed from that given in the paper by Zil’berman in the case of an isotropic spectrum of the charge carriers only by a relatively unimportant factor of order unity.

At sufficiently low temperatures that the charge carriers are scattered mainly by impurity atoms and their drift along, say, the z axis is nonzero, the asymptote of the conductivity tensor component σ_{zz} at high magnetic field has the form

$$\sigma_{zz} = - \frac{2e^3 B}{(2\pi\hbar)^2 c} \sum_{n=0}^{\infty} \int_0^{2\pi\hbar \cos \theta/a} dp_B (v_z^{nn})^2 \tau(\varepsilon_n)^{\partial f_0(\varepsilon_n)/\partial \varepsilon_n}. \quad (29)$$

Following Landau,⁶⁰ we can easily write the terms in the expression for σ_{zz} which are oscillatory on variation of the magnetic field with the aid of Poisson's formula:

$$\sum_{n=0}^{\infty} \Phi_n = \sum_{k=-\infty}^{\infty} \int_{-1/2}^{\infty} dn \Phi(n) \exp(ikn). \quad (30)$$

Here the oscillatory part of the conductivity σ_{zz}^{osc} is determined mainly by the oscillatory dependence on $1/B$ of the mean free time of the charge carriers, which is due to the summation over states of the “incoming” electrons in the collision integral.

The problem of quantum oscillations of the conductivity of metals in a magnetic field has been the subject of many papers. The most transparent and lucid derivation of the oscillatory field dependence of the elastic scattering amplitude of charge carriers on impurity atoms in the Born approximation is given in Abrikosov's monograph.²⁴ For $\hbar\omega_c \ll \eta\varepsilon_F$ the frequency of electron scattering can be written in the form

$$\frac{1}{\tau(\varepsilon)} = \frac{1}{\tau_0} (1 + \Delta_{\text{osc}}), \quad (31)$$

where

$$\Delta_{\text{osc}} = \left(\frac{e\hbar B}{m^* c \varepsilon} \right)^{1/2} \sum_e \left| \frac{\partial^2 S_e}{\partial p_B^2} \right|^{-1/2} g_e, \quad (32)$$

$$g_e = \sum_{k=1}^{\infty} a_k (-1)^k k^{-1/2} \cos\left(\frac{kcS_e}{e\hbar B} + \frac{\pi}{4} s \right) \cos\left(\frac{\pi km^*}{m} \right). \quad (33)$$

Here the a_k are numerical factors that depend on the concrete form of the carrier dispersion relation, m is the mass of a free electron, $s = \text{sgn}(\partial^2 S_e / \partial p_B^2)$, and S_e is the extremal value of the area of section of the isoenergy surface by a plane $p_B = \text{const}$. In the case of several extremal sections S_e it is necessary to sum over all possible S_e in formula (32).

As a result, the conductivity, which is proportional to the relaxation time τ , acquires an oscillatory component $\sigma^{\text{osc}} \sim \Delta_{\text{osc}} \sigma_0$.

In formulas (32) and (33) the broadening of the quantized levels of the charge carrier energy due to scattering has not been taken into account. Dingle⁷⁷ proposed that to do this it is sufficient to introduce in the oscillatory (in $1/B$) correction to the kinetic coefficient and in Δ_{osc} a factor

$$R_D = \exp(-1/\omega_c \tau), \quad (34)$$

which has come to be called the Dingle factor.

A rigorous analysis of the quantum oscillation effects, carried out by Bychkov⁷⁸ with the use of the diagram technique, showed that in many particular cases such a procedure is completely justified, although it can lead to the loss of interesting effects associated with magnetic-impurity-bound electronic states. The value calculated by Bychkov for the factor by which the amplitude of the quantum oscillations is lowered on account of scattering of conduction electrons differed from the Dingle factor only by a number factor of order

unity in the argument of the exponential factor. In this regard the Kubo method has turned out to be more attractive for studying the Shubnikov-de Haas effect. In the Kubo formalism it is unnecessary to introduce a Dingle factor in the expression for the kinetic coefficients, since the broadening of the carrier energy levels due to scattering is automatically taken into account in the description of estimation of the linear response of the electron system to the perturbation with the aid of the retarded two-time Green's functions.^{79–81}

Besides the weakening of the amplitude of the Shubnikov-de Haas oscillations σ_{zz}^{osc} due to scattering of charge carriers, there is a very significant decrease in amplitude with increasing temperature. While the part of the conductivity that changes monotonically with magnetic field, σ_{zz}^{mon} , depends weakly on temperature, since the temperature smearing of the Fermi distribution function $k_B T$ of the charge carriers is much less than the Fermi energy ε_F , the oscillatory component σ_{zz}^{osc} decreases rapidly when $k_B T$ becomes of the order of or greater than the distance $\hbar\omega_c$ between Landau levels, even for $k_B T \ll \varepsilon_F$. The factor that decreases the amplitude of the oscillations has the form

$$R_T(u) = \frac{u}{\sinh(u)}, \quad \text{where } u = \frac{2\pi^2 k_B T}{\hbar\omega_c}, \quad (35)$$

and for $u > 1$ it falls off exponentially with temperature [see formula (26)].

In the early 1950s the de Haas-van Alphen effect had already been observed in almost all metals, but for a long time the quantum oscillations of the magnetoresistance were hardly ever observed in metals for which the number of conduction electrons is of the order of one per atom. This is clearly due to the fact that the quantum correction to the classical expression for the conductivity is too small, being proportional to $(\hbar\omega_c/\varepsilon_F)^{1/2}$, while the amplitude of the quantum oscillations of the magnetic susceptibility is a factor of $(\varepsilon_F/\hbar\omega_c)^{3/2}$ larger than the Pauli paramagnetic susceptibility⁸² and the Landau diamagnetic susceptibility.⁸³

Layered organic conductors are an exceptionally convenient object for experimental study of the Shubnikov-de Haas effect, since a much larger number of charge carriers is involved in its formation than in the case of ordinary metals. In some compounds the amplitude of the resistance oscillations can reach giant values exceeding the minimum value of the resistance by one or two orders of magnitude.^{32,84–87} Clearly the theory of quantum oscillation effects developed for materials with a relatively slight anisotropy is inapplicable in this case, especially under conditions such that the distance $\hbar\omega_c$ between adjacent Landau levels is considerably greater than the width W_{\perp} of the conduction band in the direction perpendicular to the layers.

Pioneering research on the magnetic susceptibility of conductors with a markedly anisotropic quasi-two-dimensional electron energy spectrum was done in the years 1983–1985 by Wagner and co-workers^{88,89} and by Shoenberg.⁹⁰ The thermodynamic theory of quantum oscillations has been under particularly intensive development in the last 10 years (see Refs. 91–102 and the references cited therein). There is now a consistent theory of the de Haas-van Alphen effect in layered systems which can in principle be used for quantitative analysis of the magnetization oscillations.

tions observed in organic conductors (see, for example, Refs. 91 and 103–105). A number of theoretical papers have been devoted to an examination of quantum oscillations of the magnetoresistance.^{106–113} Unlike the case of the thermodynamic de Haas-van Alphen effect, the detailed description of quantum oscillations of the kinetic properties, which is complicated strongly by the necessity of taking the details of the scattering processes into account under conditions of extremely high anisotropy, is far from completed. Nevertheless, substantial progress in understanding some of the important features of the Shubnikov-de Haas effect in quasi-two-dimensional systems, at least on a qualitative level, has been made recently.

Let us consider in more detail the case of a comparatively strongly corrugated cylindrical Fermi surface, when the width of the conduction band in the direction perpendicular to the layers, W_{\perp} , is somewhat (but not very much) greater than the distance between adjacent Landau levels. This condition is realized in many layered organic conductors in fields of ~ 10 T. In this case the de Haas-van Alphen effect is still described well by the standard Lifshitz–Kosevich formula, which was obtained for moderately anisotropic metals.⁶⁴ At the same time, it turns out that the Shubnikov-de Haas effect manifests a number of specific features already at $W_{\perp} \geq \hbar \omega_c$. Among them are beats of the quantum oscillations of the magnetoresistance and an appreciable shift of their phase in relation to the beats of the oscillations of the magnetic susceptibility, which carries information about the spectrum of charge carriers.¹⁰⁹ The detection of slow quantum oscillations with a frequency proportional to the difference between the maximum and minimum areas of section of the Fermi surface has turned out to be very important.¹¹⁰ These oscillations are observed at higher temperatures than the oscillations at the fundamental frequency, which is proportional to the extremal area of section of the Fermi surface.

Figure 8 shows a typical example of the field dependence of the interlayer resistance of β -(BEDT-TTF)₂IBr₂ in a magnetic field tilted at a small angle ($\theta = 15^\circ$) from the normal to the conducting plane,¹¹⁰ at temperatures of the order of 0.6 and 1.4 K. Two types of oscillations are clearly seen. The fast oscillations, which are particularly pronounced at 0.6 K, are the Shubnikov-de Haas effect on extremal orbits of the cylindrical Fermi surface.^{29,32} By virtue of the slight corrugation of the cylinder the frequencies corresponding to the maximum and minimum cross sections of the Fermi surface are extremely close, as is reflected in the low-frequency modulation of the oscillation amplitude, which is proportional to $\cos(2\pi F_b/B + \varphi)$.

From the ratio of the beat frequency ($F_b \approx 20$ T) to the fundamental frequency ($F_0 \approx 3900$ T) one can estimate the relative value of the corrugation of the Fermi surface: $\Delta S/S = 2F_b/F_0 \approx 10^{-2}$. The ratio of the width of the conduction band in the direction perpendicular to the layers, $W_{\perp} \approx 4t_{\perp}$ [t_{\perp} is the transverse transfer integral; see formula (6)], to the distance $\hbar \omega_c$ between Landau levels in a field of 10 T is equal to $2F_b/B \approx 4$. Consequently, taking the smallness of the oscillation amplitude into account ($\sim 1\%$ of the monotonic component of the resistance) and the practically total absence of higher harmonics of the fundamental

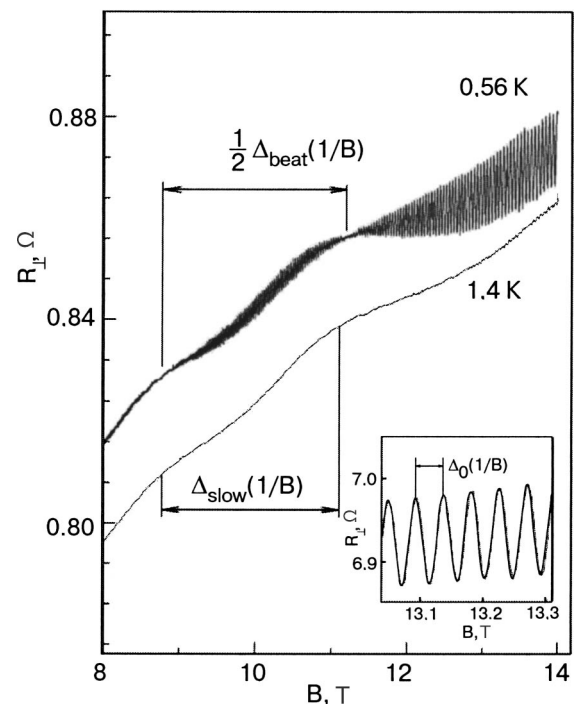


FIG. 8. Quantum oscillations of the interlayer magnetoresistance of β -(BEDT-TTF)₂IBr₂ in a magnetic field tilted at an angle $\theta \approx 15^\circ$ from the normal to the layers, for different temperatures. The fundamental Shubnikov-de Haas oscillations, shown on an enlarged scale in the inset, have a frequency $F_0 = \Delta_0^{-1}(1/B) \approx 3900$ T and beat with a frequency $F_{\text{beat}} = \Delta_{\text{beat}}^{-1}(1/B) \approx 20$ T. In addition to the fundamental oscillations, which are rapidly damped with increasing temperature, slow oscillations which are periodic on a scale of $1/B$ but are practically independent of temperature are observed. Data of Ref. 110.

frequency,^{109,110} it could be supposed that the standard “three-dimensional” model applies in the given situation. Indeed, the behavior of the magnetization oscillations in β -(BEDT-TTF)₂IBr₂ has been analyzed successfully by Wosnitza and co-workers^{31,114} in the framework of the Lifshitz-Kosevich theory.⁶⁴ However, in the case of resistance oscillations new effects arise even under these conditions, due to the quasi-two-dimensional character of the charge carriers.

Perhaps the most obvious and important anomaly of the behavior presented in Fig. 8 is the presence of slow quantum oscillations of the interlayer resistance. These oscillations, which are periodic in the $1/B$ scale, were observed back in the first experiments on this compound,^{28,29} but their nature has been understood only recently.^{110,111} It has been established¹¹⁰ that the dependence of the frequency of the slow oscillations on the orientation of the magnetic field, $F_{\text{slow}}(\theta)$, is strictly correlated with the angular oscillations of the quasiclassical part of the resistance $R_{\perp}(\theta)$, which were considered in Sec. 2.1. As is shown in Fig. 9, F_{slow} oscillates with variation of the field tilt angle θ , going to zero at angles corresponding to the maxima of $R_{\perp}(\theta)$. Such behavior attests to the direct link between the slow oscillations and the corrugation of the Fermi surface. Indeed, as was discussed above, the quasiclassical magnetoresistance takes on maximal values at those magnetic-field orientations for which the areas of all the cyclotron orbits become practically equal. This, in particular, means that the beat frequency F_b of the fundamental Shubnikov-de Haas oscillations goes to zero the

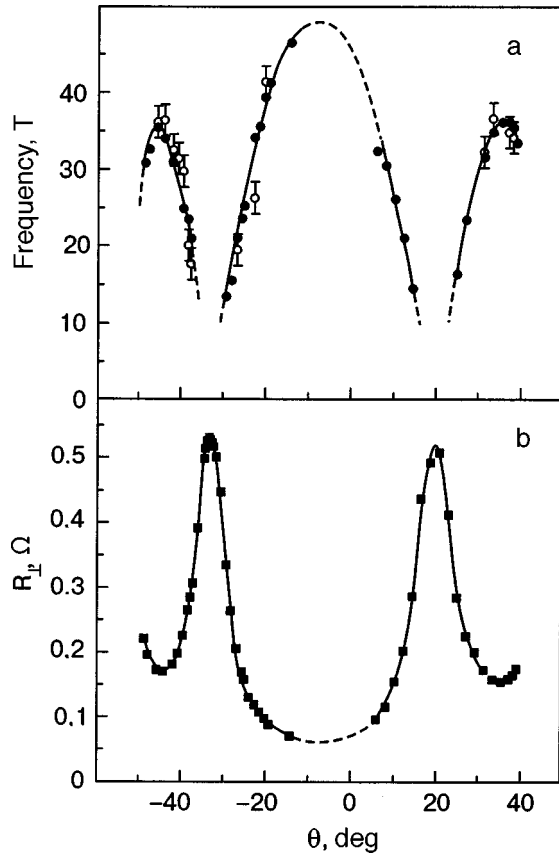


FIG. 9. Angular dependence of the frequency of slow oscillations F_{slow} (●) and twice the beat frequency $2F_{\text{beat}}$ (○) (a), and the corresponding angular dependence of the quasichlassical part of the interlayer magnetoresistance (b).¹¹⁰

the maxima of the angular oscillations of the magnetoresistance. Recent measurements have shown that the angular dependences $F_b(\theta)$ and $F_{\text{slow}}(\theta)$ actually coincide with each other to good accuracy over a wide range of angles, and the following relation holds:

$$F_{\text{slow}}(\theta) = 2F_b(\theta). \quad (36)$$

Thus one can conclude that the slow oscillations, like the beats of the fundamental Shubnikov-de Haas oscillations, are not due to some independent small section but to the weak corrugation of the main cylinder of the Fermi surface.

To explain this effect we use the model dispersion relation (6), which under conditions of quantization of the orbital motion of the electrons takes the form

$$\varepsilon(n, p_z) = \left(n + \frac{1}{2}\right) \hbar \omega_c - 2t_{\perp} \cos\left(\frac{ap_z}{\hbar}\right). \quad (37)$$

In a quantizing magnetic field \mathbf{B} deviating substantially from the layers, i.e., when $(\pi/2 - \theta) \gg \eta$, the resistance to the current transverse to the layers, as in the case when energy quantization is not taken into account, is determined mainly by the conductivity tensor component σ_{zz} .

Using the Poisson formula (30), applying it to expression (29) for σ_{zz} , we obtain a series of terms oscillatory in the inverse magnetic field:

$$\begin{aligned} \sigma_{zz} = & -\frac{4t_{\perp}^2 e^2 a}{m\hbar(2\pi\hbar)^3} \int d\varepsilon \frac{\partial f_0(\varepsilon)}{\partial \varepsilon} \tau(\varepsilon) \\ & \times \left[1 + \frac{\hbar \omega_c}{\pi t_{\perp}} \sum_{k=1}^{\infty} (-1)^k k^{-1} \cos\left(\frac{2\pi k \varepsilon}{\hbar \omega_c}\right) \right. \\ & \left. \times J_1\left(\frac{4\pi k t_{\perp}}{\hbar \omega_c}\right) R_D \right]. \end{aligned} \quad (38)$$

The mean free time of the charge carriers also oscillates with variation of the magnetic field. In the Born approximation the oscillations are determined by oscillations of the density of states and, when the carrier dispersion relation (37) is taken into account, assume the form^{115,94}

$$\tau(\varepsilon) \propto \left[1 + 2 \sum_{k=1}^{\infty} (-1)^k \cos\left(\frac{2\pi k \varepsilon}{\hbar \omega_c}\right) J_0\left(\frac{4\pi k t_{\perp}}{\hbar \omega_c}\right) R_D \right]^{-1}, \quad (39)$$

where R_D is the usual Dingle factor, and $J_0(u)$ and $J_1(u)$ are Bessel functions. As is seen from expressions (38) and (39), the oscillations of the relaxation time and velocity of the electrons are modulated in amplitude by the Bessel functions of order zero and one, respectively, with an argument determined, as expected, by the ratio $t_{\perp}/(\hbar \omega_c)$. Obviously the product of two oscillatory functions in the expression for the conductivity will contain a slowly oscillating term.

At large arguments $u = 4\pi t_{\perp}/(\hbar \omega_c)$ one can use the approximate expressions for the Bessel functions $J_0(u) \approx \sqrt{2/\pi u} \cos(u - \pi/4)$ and $J_1(u) \approx \sqrt{2/\pi u} \sin(u - \pi/4)$. Since in the experiment the amplitude of the oscillations is rather small, and all the harmonics except the first are substantially suppressed, we neglect the oscillations of the chemical potential and keep in the expansion for σ_{zz} only the terms of lowest order in the factors R_D and R_T , and after integration over ε we obtain¹¹⁰

$$\begin{aligned} \sigma_{zz} = & \sigma_0 \left(\frac{t_{\perp}}{\varepsilon_F}\right)^2 \left\{ 1 + 2 \sqrt{\frac{\hbar \omega_c (1 + a^2)}{2\pi^2 t_{\perp}}} \cos\left(\frac{2\pi \mu}{\hbar \omega_c}\right) \right. \\ & \times \cos\left(\frac{4\pi t_{\perp}}{\hbar \omega_c} - \frac{\pi}{4} + \varphi\right) R_D R_T + \frac{\hbar \omega_c}{2\pi^2 t_{\perp}} \\ & \left. \times R_D^{*2} \sqrt{1 + a^2} \cos\left(\frac{8\pi t_{\perp}}{\hbar \omega_c} - \frac{\pi}{2} + \varphi\right) \right\}, \end{aligned} \quad (40)$$

where

$$a = \hbar \omega_c / (2\pi t_{\perp}), \quad \varphi = \arctan(a), \quad (41)$$

μ is the chemical potential of the charge carriers, and σ_0 is the monotonic part of the conductivity along the layers.

The second term in the sum (40) corresponds to the main oscillations with the fundamental frequency $F_0 = \mu m^* c / \hbar e = cS/(2\pi\hbar e)$, modulated in amplitude at the frequency $F_{\text{slow}} = 4t_{\perp}/\hbar \omega_c = 2t_{\perp} m^* c / \hbar e$. The third term in (40) corresponds to slow oscillations with a frequency equal to twice the beat frequency, in complete agreement with experiment.

It must be noted that the amplitude of the slow oscillations does not contain a temperature factor, since these oscillations are independent of the electron energy (they are determined only by the value of t_{\perp} in the dispersion relation). Indeed, as is seen in Fig. 8, the amplitude of the slow oscil-

lations remains almost unchanged when the temperature is increased from 0.6 to 1.4 K, while the fundamental harmonic of the Shubnikov oscillations is almost completely suppressed at 1.4 K. Strictly speaking, the slow oscillations are, of course, damped with temperature by virtue of the temperature-dependent scattering processes,¹¹¹ but this damping is considerably slower. In particular, in the compound β -(BEDT-TTF)₂IBr₂ the slow oscillations are observed all the way up to temperatures ~ 10 K, i.e., an order of magnitude higher than the fundamental Shubnikov oscillations.

Another consequence of the fact that the slow oscillations are independent of the carrier energy is the difference of the corresponding Dingle factor R_D^* from the standard one.^{110,111} Usually the relaxation time that enters into the Dingle factor takes into account both point defects and other, macroscopic, imperfections of the crystal which influence the local value of the chemical potential: dislocations, mosaicity, local strains, etc. Since the chemical potential does not appear in the expression for the slow oscillations, the factor R_D^* is determined solely by the point defects. Indeed, experiment¹¹⁰ has shown that in β -(BEDT-TTF)₂IBr₂ the relaxation time obtained from the Dingle factor of the slow oscillations is five times larger than the value determined from the fundamental Shubnikov-de Haas oscillations. Hence it can be concluded that the contribution of the macroscopic inhomogeneities to the suppression of the Shubnikov oscillations is dominant in this compound.

Thus the slow oscillations of the interlayer magnetoresistance are a general phenomenon observed in sufficiently pure layered metals by virtue of the superposition of the oscillations of the relaxation time and carrier velocity in the direction transverse to the layers, the amplitudes of which turn out to be comparable, when W_\perp is of the order of $\hbar\omega_c$. Indeed, such oscillations have been observed not only in β -(BEDT-TTF)₂IBr₂ but also in a number of other layered organic conductors in fields ~ 10 T.^{116–120}

It follows from relations (40) and (41) that the phase of the beats of the fundamental harmonic of the Shubnikov-de Haas oscillations contains an anomalous term φ which depends on the magnetic field. In fact, experiments on a number of organic metals^{109,121–123} have revealed a significant phase shift of the beats of the magnetoresistance oscillations with respect to the beats of the magnetization oscillations in fields of the order of 10–20 T (recall that the latter are well described by the standard three-dimensional Lifshitz-Kosevich model⁶⁴).

A detailed experimental study of the phase of the beats of the Shubnikov oscillations was carried out in Refs. 109 and 123, and it was shown that it is indeed substantially dependent on the ratio $\hbar\omega_c/W_\perp$. In particular, it was found that the phase shift φ increases strongly as the magnetic-field orientation approaches the direction corresponding to the peak in the angular oscillations of the classical part of the magnetoresistance. This result agrees with the fact that the effective interlayer transfer integral vanishes at the peak of the angular oscillations.¹²⁴ It should be noted, however, that the phase shift measured in Ref. 109 was approximately twice as large as the value predicted by formulas (40). This is illustrated in Fig. 10, where the symbols correspond to the

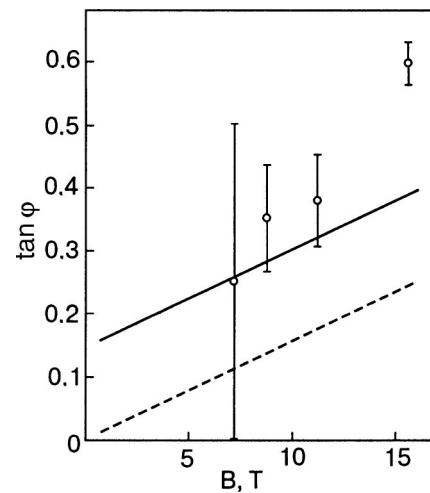


FIG. 10. Tangent of the phase shift of the beats of the Shubnikov oscillations in β -(BEDT-TTF)₂IBr₂ as a function of magnetic field according to the data of Ref. 109. The dashed line was constructed according to formulas (41), and the solid line is the result of a quantum-mechanical calculation.¹¹¹ The calculation was done using the values of the cyclotron mass $m_c = 3.9 \times 10^{-27}$ g and scattering time on point defects $\tau^* = 8 \times 10^{-12}$ s obtained from the Dingle factor of the slow oscillations.¹¹⁰

results of the measurements and the dashed line represents relation (41); here the ratio $\hbar\omega_c/t_\perp$ was taken from the beat frequency of the de Haas-van Alphen oscillations.

Subsequently a more rigorous theoretical analysis of the conductivity based on the Kubo formalism with the use of the self-consistent Born approximation¹¹¹ showed that a term $\approx \hbar/(2t_\perp\tau^*)$ must be added to the argument of the arctangent in formula (41), where τ^* is the scattering time on point defects (it can be determined from the Dingle factor for the slow oscillations, R_D^*). Then, as is shown in Fig. 10 (solid line), fair agreement with experiment¹⁰⁹ can be achieved for fields in the interval 7–12 T, which corresponds to values $\hbar\omega_c/t_\perp \approx 0.7$ –1.2. However, upon further increase of the field the discrepancy between the theory¹¹¹ and experiment increases. Moreover, it has been observed¹²³ that for field orientations close to the direction corresponding to the peak in the angular oscillations of the magnetoresistance the phase shift φ is significantly greater than the limit $\pi/2$ predicted by the theory.¹¹¹

Thus the results presented show that, unlike the thermodynamic quantum oscillations such as the de Haas-van Alphen effect, the quantum oscillations of the conductivity of layered metals display a number of anomalies even for $\hbar\omega_c/W_\perp < 1$. The existing theoretical models are able to describe the nature of these anomalies in a qualitative way. However, to achieve quantitative agreement between theory and experiment, especially under conditions when the ratio $\hbar\omega_c/W_\perp$ increases, approaching unity, further efforts will be necessary.

For $\hbar\omega_c \gg W_\perp$ the Boltzmann transport equation does not lead to a satisfactory result at high magnetic fields ($\omega_c\tau \gg 1$). A detailed analysis of the conductivity σ_{zz} for the case $\hbar\omega_c \gg W_\perp$ in a field perpendicular to the layers ($\mathbf{B} \parallel \mathbf{z}$) was done by Champel and Mineev¹¹² and by Gvozdkov¹¹³ on the basis of the Kubo formalism with the use of the self-consistent Born approximation. It was shown that with increasing field an ever greater contribution to the oscillations

of σ_{zz} is given by the purely quantum term, which does not have an analog in the semiclassical model. In particular, at integer occupation of the Landau levels this term almost completely compensates the semiclassical Boltzmann contribution, which gives rise to a “pseudogap” in the function $\sigma_{zz}(\varepsilon)$ and an activation temperature dependence $\sigma_{zz}(T)$ at integer $\mu/\hbar\omega_c$. Such a temperature dependence has indeed been observed¹²⁵ at fields greater than 20 T in the organic conductor β'' -(BEDT-TTF)₂SF₅CH₂CF₂SO₃. Nevertheless, a detailed comparison of the Shubnikov–de Haas oscillations in this compound^{125,126} with the results of the theoretical calculations reveals significant discrepancies. As was noted in Ref. 112, the cause of this may be the insufficiency of taking into account scattering only on point defects in the regime $\hbar\omega_c \gg W_{\perp}$. A more rigorous treatment of scattering processes in layered organic conductors is an extremely complex problem and has not been done at the present time.

4. HALL EFFECT

The Hall field at high magnetic fields, even in the presence of open sections of a Fermi surface in the form of a corrugated cylinder, i.e., for $\theta = \pi/2$, has the form⁴⁸

$$\mathbf{E}_{\text{Hall}} = \frac{[\mathbf{j} \times \mathbf{B}]}{Nec}, \quad (42)$$

i.e., the same form as in the case of a magnetic field tilted with respect to the layers, when all the charge carriers in the collisionless limit ($\tau = \infty$) drift at a velocity

$$\mathbf{u} = c \frac{[\mathbf{E} \times \mathbf{B}]}{B^2}. \quad (43)$$

This is because the drift of the charge carriers along open orbits in a plane orthogonal to the magnetic field, with the velocity

$$v_x = \frac{2\pi\hbar c}{aeBT_B(p_B)}, \quad (44)$$

is naturally compensated in the expression for the current density

$$\mathbf{j} = Ne\mathbf{u}, \quad (45)$$

where \mathbf{u} , as in the case when θ is not equal to $\pi/2$, has the form (43). As a result, by measuring the value of the Hall field at a high magnetic field with any orientation, one can determine the charge carrier density N to the necessary accuracy.

At low temperatures such that it is important to take the quantization of the electron energy levels into account, for conductors with a single group of charge carriers the quantum corrections to the asymptote of the Hall field (42) for $\omega_c\tau \gg 1$ appear only in the higher terms of the expansion in a power series in the small parameter $\gamma = 1/\omega_c\tau$. Let us present Lifshitz' elegant proof of this assertion.⁷⁴

We calculate the current density flowing in the direction orthogonal to the magnetic field with the aid of the solution of the quantum kinetic equation for the statistical operator $\hat{f} = \hat{f}_0 + \hat{f}_1$:

$$\{H\hat{f}\} = \hat{W}_{\text{coll}}(\hat{f}), \quad (46)$$

where $H = H_0(\mathbf{P} - e\mathbf{A}/c) - e\mathbf{E} \cdot \mathbf{r}$ is the Hamiltonian of a conduction electron in uniform electric and magnetic fields, \mathbf{P} is the generalized momentum operator, \mathbf{A} is the vector potential for the magnetic induction $\mathbf{B} = (0, B \sin \theta, B \cos \theta) = \text{curl} \mathbf{A}$, which in the Landau gauge has the form $\mathbf{A} = (0, xB \cos \theta, -xB \sin \theta)$, and in the absence of electric field the projections of the generalized momentum P_y and P_z are good quantum numbers.

The quantum analog of the collision integral $\hat{W}_{\text{coll}}(\hat{f})$ takes into account the scattering of electrons by the potential $\sum_i V(\mathbf{r} - \mathbf{r}_i)$ of impurity atoms located at coordinates \mathbf{r}_i . In the case when this potential is short-ranged and weak, the collision integral \hat{W}_{coll} can lead to an integral operator acting on a single-particle statistical operator $\hat{f} = \hat{f}_0 + \hat{f}_1$. The operator \hat{f}_0 describes the unperturbed state of the system of conduction electrons, —its diagonal matrix elements are equal to the Fermi distribution function of the charge carriers, $f_0^{nn}(p_B) = f_0\{\varepsilon_n(p_B)\}$ —and the operator \hat{f}_1 describes the perturbation of the charge carriers by the electric field.

In an approximation linear in the weak electric field the kinetic equation takes the form

$$\{H_0\hat{f}_1\} - \{e\mathbf{E} \cdot \mathbf{r}\hat{f}_0\} = \hat{W}_{\text{coll}}\{\hat{f}_1\}. \quad (47)$$

It is easy to see that the expression for the current density component orthogonal to the magnetic field,

$$[\mathbf{j} \times \mathbf{B}]/B = \text{Tr}(e[\hat{\mathbf{v}} \times \mathbf{B}]\hat{f}_1)/B, \quad (48)$$

where $\hat{\mathbf{v}}$ is the velocity operator of the conduction electrons, is proportional to the change of the momentum of the electron with time:

$$e[\hat{\mathbf{v}} \times \mathbf{B}] = c \frac{d}{dt} \hat{\mathbf{p}} = c\{H, \hat{\mathbf{p}}\}. \quad (49)$$

Using relation (49), we obtain

$$\begin{aligned} [\mathbf{j} \times \mathbf{B}]_y &= (ic/h) \text{Tr}(H\hat{f}_1\hat{p}_y - \hat{f}_1H\hat{p}_y) \\ &= -c \text{Tr}(\{e\mathbf{E} \cdot \mathbf{r}\hat{f}_0\}\hat{p}_y) - c \text{Tr}(\hat{W}_{\text{coll}}(\hat{f}_1)\hat{p}_y). \end{aligned} \quad (50)$$

As a result, in the collisionless limit $\hat{W}_{\text{coll}}(\hat{f}_1) = 0$ we obtain

$$[\mathbf{j} \times \mathbf{B}]_y = ce \text{Tr}(E_y\hat{f}) = \text{Tr}(\hat{f}_0)ecE_y = NecE_y, \quad (51)$$

i.e., at arbitrary orientations of the current density and magnetic field the asymptote of the Hall field has the form (42). Taking the collision integral into account is extremely important for calculating the dissipative component of the current density.

Thus in layered conductors with a Fermi surface in the form of a slightly corrugated cylinder, the quantum oscillations of the off-diagonal components of the magnetoresistivity tensor, divided by the asymptote of the Hall field, are smaller than the quantum oscillations $\rho^{\text{osc}}/\rho^{\text{mon}}$ by at least a factor of $\omega_c\tau$.

However, in conductors with a multisheet Fermi surface the amplitude of the oscillations of the Hall field can be comparable to the the amplitude of the magnetoresistance oscillations.

Let us consider the case when the Fermi surface includes a pair of slightly corrugated planes in addition to the cylin-

dricul part. Such a topology of the Fermi surface is quite common in organic metals.^{31–33} In particular, it is characteristic for one of the best-known organic superconductors, κ -(BEDT-TTF)₂Cu(NCS)₂ (Ref. 30) and also for the normal metallic state of the compound α -(BEDT-TTF)₂MHg(SCN)₄, where M is a metal from the group K, Rb, Tl or NH₄ (Ref. 127). Open sections of such a Fermi surface by a plane $p_B = \text{const}$ are encountered at practically any orientation of the magnetic field, and the magnetoresistance of such a conductor increases without saturation as the magnetic field is increased. The position of all the planes in p space can easily be determined from the anisotropy of the magnetoresistance in the plane of the layers.²⁰

For $\eta \ll \cos \theta$ and $\gamma_0 \ll \cos \theta$ the magnetoresistance to a current flowing along the layers has the form^{49,128}

$$\begin{aligned} \rho_{xx} &= \frac{\sigma_1 \sin^2 \varphi \cos^2 \theta + \gamma_0^2 \sigma_0}{\gamma_0^2 \sigma_0 (\sigma_0 + \sigma_1)}; \\ \rho_{yy} &= \frac{\sigma_1 \cos^2 \varphi \cos^2 \theta + \gamma_0^2 \sigma_0}{\gamma_0^2 \sigma_0 (\sigma_0 + \sigma_1)}, \end{aligned} \quad (52)$$

where σ_0 and σ_1 are equal in order of magnitude to the contribution from the charge carriers situated, respectively, on the cylindrical and planar parts of the Fermi surface to the conductivity along the layers in the absence of magnetic field and φ is the angle between the p_y axis and the corrugated plane of the Fermi surface.

The contribution of the charge carriers whose states belong to a sheet of the Fermi surface in the form of a slightly corrugated plane to the conductivity along the normal to the layers does not lead to a noticeable Hall effect provided that $\eta \ll \gamma_0$ and the resistivity along the z axis,

$$\rho_{zz} = \frac{1}{\sigma_{zz}} + \frac{\sigma_1 \cos^2 \varphi \sin^2 \theta}{\gamma_0^2 \sigma_0 (\sigma_0 + \sigma_1)} \quad (53)$$

is determined mainly by the conductivity tensor component σ_{zz} .

In formula (52) and in the last term of formula (53) we have dropped unimportant factors of order unity which depend on the concrete form of the dispersion relation of the charge carriers, and the relationship between the mobilities of the conduction electrons of the two groups are assumed to be the same.

At rather high magnetic fields, when $\gamma_0 \leq \eta$, the Hall components of the resistivity tensor, in particular,

$$\rho_{xz} = \gamma_0^{-2} \frac{\sigma_1 \sin 2\varphi \sin 2\theta}{\sigma_0 (\sigma_0 + \sigma_1)} + \gamma_0^{-1} \frac{(\sigma_0 + \sigma_1 \sin^2 \varphi) \sin \theta}{\sigma_0 (\sigma_0 + \sigma_1)} \quad (54)$$

are already comparable to ρ_{zz} .

The presence of an additional pocket of the Fermi surface in the form of weakly corrugated planes leads for $\gamma_0 \leq \eta$ to the unrestricted growth of the resistivity to a current transverse to the layers with magnetic field, and the Hall field, which is proportional to H^2 , is now comparable to E_z .

The energy spectrum of the carriers whose states belong to the slightly corrugated planes in momentum space does not contain discrete levels, and therefore this group of conduction electrons does not take part in the formation of quantum oscillation effects, but its presence can have a substantial

influence on the asymptote of the quantum oscillations of the Hall field. When the oscillatory dependence of the mean free time on $1/B$ at low temperatures for the conduction electrons with closed orbits on a sheet of the Fermi surface in the form of a corrugated cylinder is taken into account, the Hall field in a quantizing magnetic field takes the form¹²⁸

$$\begin{aligned} E_x &= j \frac{H \sin \theta}{2Nec} \\ &\times \left\{ -\sin^2 \varphi + \frac{H\sigma_0\sigma_1 \sin 2\varphi \cos \theta - Nec\sigma_0 \cos^2 \varphi}{Nec[\sigma_0 + \sigma_1(1 + \Delta_{\text{osc}})]} \right\}, \end{aligned} \quad (55)$$

$$E_y = j \frac{H \sin \theta}{Nec} \frac{H\sigma_0\sigma_1 \cos \theta - Nec\sigma_1(1 + \Delta_{\text{osc}}) \sin 2\varphi}{Nec[\sigma_0 + \sigma_1(1 + \Delta_{\text{osc}})]}. \quad (56)$$

It is easy to see that the ratio of the oscillatory part $E_{\text{Hall}}^{\text{osc}}$ of the Hall field to the part that varies monotonically with magnetic field, $E_{\text{Hall}}^{\text{mon}}$, as follows from formulas (55) and (56),

$$E_{\text{Hall}}^{\text{osc}}/E_{\text{Hall}}^{\text{mon}} = -\Delta_{\text{osc}} \frac{\sigma_1}{\sigma_0 + \sigma_1} \quad (57)$$

is of the same order of magnitude as $\rho_{zz}^{\text{osc}}/\rho_{zz}^{\text{mon}}$.

Thus, by studying the dependence of the resistivity and Hall field on the value of a sufficiently strong magnetic field for different orientations of the field with respect to the layers, one can reconstruct completely the topological structure of the Fermi surface and determine the contribution of the individual groups of charge carriers to the total conductivity of the conductor.

5. CONCLUSION

At high magnetic fields the kinetic characteristics of low-dimensional conductors depend substantially on the concrete form of the energy spectrum of the charge carriers and contain detailed information about the dispersion relation of the conduction electrons. Intensive research on galvanomagnetic phenomena in organic layered conductors at low temperatures has revealed a number of peculiar effects specific to quasi-two-dimensional conductors. It turns out that the electronic phenomena in quasi-two-dimensional conductors are manifested in fundamentally different ways for different orientations of the quantizing magnetic field. Even in a moderately strong magnetic field orthogonal to the layers, when the discrete-continuous electron energy spectrum of the layered conductor contains a rather large number of quantized values of the momentum projection on the magnetic field direction at a fixed value of the energy, e.g., equal to the Fermi energy, at certain orientations of the magnetic field with respect to the layers only one or a few electron states with the Fermi energy are possible, i.e., the electron energy spectrum turns out to be quasi-discrete.

Thus, depending on the angle between the magnetic field vector and the normal to the layers, a layered conductor behaves as a three-dimensional conductor with highly anisotropic electron energy spectrum or as a two-dimensional conductor with a discrete spectrum of charge carriers. This orientation effect, which is specific to quasi-two-dimensional

conductors, is manifested in various kinetic phenomena. In particular, for certain orientations of the magnetic field a layered conductor is transparent to electromagnetic and acoustic waves,^{129,130} and in such conductors the propagation of peculiar weakly damped electromagnetic and spin waves can occur,^{131,132} carrying information about the electron energy spectrum and the relaxation properties of the charge carriers.

Layered conductors with a quasi-two-dimensional electron energy spectrum are extremely convenient objects for studying quantum oscillation effects, the formation of which involves a large number of charge carriers. The detection of low-frequency oscillations of the magnetoresistance of layered conductors at high temperatures such that the fundamental harmonics of the Shubnikov-de Haas oscillations are hardly observed, and the study of the phase relations of the oscillations of the magnetoresistance and magnetic susceptibility can yield detailed information about the dispersion relation of the conduction electrons.

Linear growth of the resistivity with magnetic field (the Kapitsa law) is very specific in quasi-two-dimensional conductors, being observed even in single-crystal samples. Linear growth of the resistivity of bismuth with magnetic field, which gave way to quadratic growth at higher fields, was observed by E. S. Borovik,¹³³ who devoted many years to studying the galvanomagnetic phenomena in metals. The magnetoresistance of a layered conductor in a magnetic field lying in the plane of the layers behaves in an analogous way. It is quite possible that the linear growth of the resistivity of bismuth with magnetic field is due to the marked anisotropy of the Fermi surface of that substance, the axes of the electron ellipsoid of the Fermi surface of bismuth differing by roughly a factor of 10.

The investigation of galvanomagnetic phenomena in layered conductors has been the subject of an enormous number of studies. We have not attempted to cover everything, and some interesting results of experimental and theoretical research on the classical and quantum galvanomagnetic effects may have been left out. The main goal of this paper was to demonstrate the effectiveness of galvanomagnetic measurements for investigating the electron energy spectrum.

We are grateful to the Editor-in-Chief of the journal *Low Temperature Physics* for inviting us to contribute to this special issue dedicated to the 90th anniversary of the birth of the outstanding physicist and fascinating man, Evgenii Stanislavovich Borovik.

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