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## Angular magnetoresistance oscillations and the shape of the Fermi surface in $\beta$ -(ET)<sub>2</sub>IBr<sub>2</sub>

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**Abstract.** — Angular magnetoresistance oscillations have been studied systematically for  $\beta$ -(ET)<sub>2</sub>IBr<sub>2</sub> in the magnetic field rotating in a series of planes perpendicular to the conducting (a, b)-plane. The oscillations have been found in all studied planes. The shape of the Fermi surface transverse cross-section has been reconstructed using the obtained data. Angular dependence of the slow Shubnikov-de Haas oscillations frequency and some fine features of angular magnetoresistance oscillations permit to discuss also the structure of the Fermi surface longitudinal cross-section. The Fermi surface consists most likely of main cylinders with inclined warping planes and small pockets or necks between them.

### 1. Introduction.

Recently, strong angular magnetoresistance oscillations, occurring upon rotating the magnetic field in a plane perpendicular to the conducting layers, have been discovered [1 – 5] in a number of layered organic metals. The magnetoresistance was found to reach its local maxima periodically in  $\cot \varphi$ , where  $\varphi$  is the angle between the field direction and the conducting (a, b)-plane. The first explanation of the phenomenon [6] employed the fact that the distribution of the cyclotron orbit areas for a Fermi surface in the form of a weakly corrugated cylinder degenerates periodically in  $\cot \varphi$  upon magnetic field rotation. It was supposed that the degeneration

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should result in increasing the magnetoresistance. Later, the angular magnetoresistance oscillations were explicitly obtained in resistivity tensor calculations [7] (see also [8]) for a quasi two-dimensional metal.

An alternative explanation of the magnetoresistance increase for some field directions was proposed in [2, 8] as resulting from the existence of self-crossing trajectories on the Fermi surface. In the case of a weakly corrugated cylindrical Fermi surface the self-crossing trajectories may be associated, for example, with the existence of some necks connecting adjacent cylinders. In this situation the oscillations should disappear for some planes of magnetic field rotation. So, a possible way to distinguish the different explanations might consist in a systematic investigation of angular magnetoresistance oscillations upon rotating magnetic field in a series of planes perpendicular to the conducting plane.

We have performed such measurements on a number of high quality single crystals of  $\beta$ -(ET)<sub>2</sub>IBr<sub>2</sub>. Although the oscillations amplitude have been found to vary for different planes of the field rotation, they do not disappear for any plane. Therefore, we conclude that the angular oscillations are caused mainly by the first of the two above mentioned mechanisms. Considerable enhancement of the Shubnikov-de Haas (SdH) oscillations amplitude observed for the field directions corresponding to local maxima of the magnetoresistance [1, 2] also supports this conclusion [6]. Nevertheless, basing on our results the existence of a more complicated Fermi surface than a simple warped cylinder cannot be completely excluded.

A substantial result of our investigations is the reconstruction of the Fermi surface cross-section shape and dimensions in the (a, b)- plane. The possibility of such a reconstruction on the basis of numerical calculations has been pointed out in [9]. We have used, however, for this purpose an explicit analytic expression derived in [9] which relates the magnetoresistance maxima angles with the maximum projection of the Fermi momentum vector on the direction of the magnetic field component parallel to the (a, b)-plane.

In section 2 we discuss the physical reasons for the angular magnetoresistance oscillations and refine some theoretical ideas concerning their explanation. In section 3 experimental results of the magnetoresistance measurements for different field rotation planes are presented and the form of the Fermi surface is reconstructed.

## 2. Theoretical considerations.

Let us consider a quasi-two-dimensional metal with the following electron dispersion law:

$$E(p_x, p_y, p_z) = \varepsilon(p_x, p_y) - 2t \cos([p_z d + p_x u_x + p_y u_y]/\hbar). \quad (1)$$

Here  $(p_x, p_y) \equiv \mathbf{p}_{\parallel}$  and  $p_z$  are the momentum components parallel and perpendicular to the conducting planes;  $t$  and  $d$  are, respectively, the hopping integral and the spacing between adjacent conducting planes;  $\varepsilon(p_x, p_y) = \varepsilon(-p_x, -p_y)$  is an in-plane dispersion law. Equation (1) is written under the assumption that electrons hop between the conducting planes along vectors  $\mathbf{h}$  and  $-\mathbf{h}$  in the  $(x, y, z)$  space,  $\mathbf{h} = (u_x, u_y, d)$ ,  $d > 0$ . We shall consider the case

$$\eta \equiv t/\varepsilon_F \ll 1 \quad (2)$$

( $\varepsilon_F$  is the Fermi energy), so that the Fermi surface is a weakly corrugated cylinder with a closed convex curve of an arbitrary form in a cross-section. The cylinder axis is parallel to  $z$ -axis, while the planes of warping are inclined with respect to the  $(x, y)$ -plane being perpendicular to the hopping vector  $\mathbf{h}$ .

Magnetic field direction is determined by two angles,  $\varphi$  and  $\theta$ , according to the formula  $\mathbf{H} = (H \cos \theta \cos \varphi, H \sin \theta \cos \varphi, H \sin \theta)$ . Let us consider the case of strong magnetic field:  $\gamma \equiv 1/\omega_c \tau \ll 1$ , where  $\omega_c$  is the cyclotron frequency,  $\tau$  the relaxation time. Electrons move along the orbits in the planes perpendicular to  $\mathbf{H}$ ; each orbit may be labeled by a coordinate,  $P_z$ , of the point where its plane intersects the  $p_z$ -axis. For generic values of  $\varphi \neq 0$  and  $P_z$  the mean velocity component parallel to the field direction,  $\bar{v}_H(P_z)$ , is non-zero, whereas the mean perpendicular components are zero. The bar over  $v$  means the averaging over the electron classical motion along the orbit.

In the system of coordinates, related to the field direction, the diagonal component of the conductivity tensor along the magnetic field,  $\sigma_{HH}$ , may be written as [10]

$$\sigma_{HH} = \sigma_{HH}^{(0)} + O(\gamma^2), \quad (3)$$

where

$$\sigma_{HH}^{(0)} \propto \langle \bar{v}_H^2 \rangle \quad (4)$$

and the angular brackets mean the averaging over  $P_z$ . All other components of the conductivity tensor go to zero as  $\gamma$  or  $\gamma^2$  [10]. Transformation to the system of coordinates related to the crystal leads to the following form of the conductivity tensor (see [8])

$$\sigma_{ij} = \begin{pmatrix} O(\gamma^2) & O(\gamma) & O(\eta^2 \gamma) \\ -O(\gamma) & \sigma_{zz}^{(0)} \cot^2 \varphi + O(\gamma^2) & \sigma_{zz}^{(0)} \cot \varphi + O(\eta^2 \gamma) \\ -O(\eta^2 \gamma) & \sigma_{zz}^{(0)} \cot \varphi - O(\eta^2 \gamma) & \sigma_{zz}^{(0)} + O(\eta^2 \gamma^2) \end{pmatrix}, \quad i, j = x, y, z, \quad (5)$$

where

$$\sigma_{zz}^{(0)} = \sigma_{HH}^{(0)} \sin^2 \varphi \propto \langle \bar{v}_z^2 \rangle. \quad (6)$$

Inversion of tensor (5) under condition (2) gives the following expressions for the resistivity tensor components

$$\rho_{zz} = 1/\sigma_{zz}, \quad \rho_{xx} \sim \rho_{yy} = \rho_0 + [O(\eta^4) + O(\eta^2 \gamma^2)] \sigma_{zz}^{-1}, \quad (7)$$

where  $\rho_0$  is of the order of the in-plane resistivity without magnetic field and

$$\sigma_{zz} = \sigma_{zz}^{(0)} + O(\eta^2 \gamma^2). \quad (8)$$

The origin of the angular oscillations may be explained now as follows. When magnetic field has a generic direction,  $\bar{v}_z(P_z) = \bar{v}_H(P_z) \sin \varphi$  is non-zero. So, with increasing magnetic field the resistivity grows and reaches the saturation value given by equation (7) with  $\sigma_{zz} = \sigma_{zz}^{(0)}$ , where  $\sigma_{zz}^{(0)}$  is determined by equation (6). But, as it will be shown below, there exist special field directions at which  $\bar{v}_z(P_z)$  nearly vanishes for all values of  $P_z$ . For these special directions the second term in equation (8) plays the main role and resistivity (7) grows as  $H^2$  to saturate at very high field beyond experimental capabilities. Thus, when the constant magnetic field is rotated, the resistivity exhibits strong maxima at these particular directions. This result was firstly obtained in [7], basing on numerical and analytical calculations, and then was discussed also in [8].

It follows from equation (7) that all diagonal components of the resistivity tensor contain  $\sigma_{zz}$  in denominator and thus should exhibit the angular oscillations, although  $\rho_{xx}$  and  $\rho_{yy}$  contain  $\sigma_{zz}^{-1}$  with a small coefficient. This is result of a general Boltzmann equation approach. However, in the  $\tau$ -approximation for the parabolic dispersion law  $\varepsilon(p_x, p_y)$  it was found both

numerically [7] and analytically [8] that the nominator exactly cancels the denominator in equation (7), so that  $\rho_{xx}$  and  $\rho_{yy}$  do not exhibit angular oscillations. Experimentally, all the components of the resistivity tensor exhibit strong oscillations [1 – 3].

The values of the critical angles were found analytically [6] for a model dispersion law and numerically [9] for the dispersion law calculated in [11]. Below we describe the general analytical results of [8]. According to (1) the mean velocity  $\bar{v}_z$  can be written as

$$\bar{v}_z = \overline{\partial E / \partial p_z} = \frac{2td}{T\hbar} \int_0^T d\xi \sin \{ [p_z(\xi)d + (\mathbf{p}_{\parallel}(\xi)\mathbf{u})] / \hbar \}, \quad (9)$$

where  $\xi$  represents the time variable,  $T$  is the period of rotation, the time dependence  $\mathbf{p}(\xi)$  has to be determined from classical equations of motion with Lorentz force. When an electron moves in the plane tilted at the angle  $\varphi$  with respect to the  $(x, y)$  plane, the following relation holds:

$$p_z(\xi) = P_z - p_H(\xi) \cot \varphi, \quad (10)$$

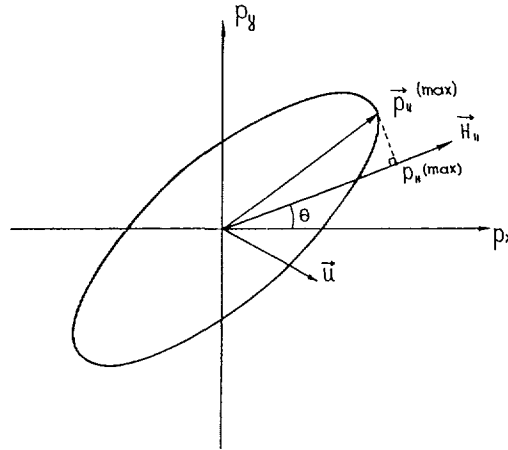


Fig. 1. — Schematic look at the transverse cross-section of a Fermi surface.  $\mathbf{H}_{\parallel}$  is the magnetic field component parallel to the  $(x, y)$ -plane,  $\mathbf{p}_{\parallel}^{(\max)}$  is the in-plane component of the Fermi momentum whose projection on the direction of  $\mathbf{H}_{\parallel}$  attains the maximal value  $p_H^{(\max)}$ ,  $\mathbf{u}$  is the in-plane part of the hopping vector.

where  $P_z$  labels the position of the plane and  $p_H$  is the projection of the  $\mathbf{p}_{\parallel}$  vector on the direction of the in-plane component of the magnetic field (see Fig.1). Following [8] let us consider relatively small  $\varphi$ , where  $\cot \varphi \gg 1$  and the integrand in (9) oscillates rapidly due to relation (10). The integral can be taken in the vicinity of the two points,  $\mathbf{p}_{\parallel}^{(\max)} = -\mathbf{p}_{\parallel}^{(\min)}$ , where  $p_H(\xi)$  takes the maximum,  $p_H^{(\max)} > 0$ , and the minimum,  $-p_H^{(\max)} < 0$ , values, respectively, (see Fig.1):

$$\bar{v}_z(P_z) \propto \sin \left( \frac{P_z d}{\hbar} \right) \cos \left( \frac{|dp_H^{(\max)} \cot \varphi - (\mathbf{p}_{\parallel}^{(\max)} \mathbf{u})|}{\hbar} - \frac{\pi}{4} \right). \quad (11)$$

It follows from (11) that the critical angles, where  $\bar{v}_z(P_z) = 0$ , obey the relation

$$|\cot \varphi_c| = [\pi \hbar (N - 1/4) \pm (\mathbf{p}_{\parallel}^{(\max)} \mathbf{u})] / p_H^{(\max)} d, \quad (12)$$

where  $N$  is an integer, the signs  $\pm$  correspond to positive and negative values of  $\varphi_c$ , respectively. Relation (12) permits to determine experimentally the form of the Fermi surface cross-section by rotating magnetic field in different planes perpendicular to the  $(x, y)$ -plane. For every value of  $\theta$  one can deduce  $p_H^{(\max)}(\theta)$  from the slope of the  $|\cot \varphi_c|$  vs.  $N$  linear dependence (12). Then it is necessary to put a point at the distance  $p_H^{(\max)}$  from the origin in the direction given by the corresponding angle  $\theta$  and draw a line through this point in the perpendicular direction (see Fig.1). The envelope of the lines drawn for all angles  $\theta$  gives the Fermi surface cross-section.

It follows from (12) that for a given  $N$  the values of  $|\cot \varphi_c|$  for positive and negative  $\varphi_c$  differ by  $2(\mathbf{p}_{\parallel}^{(\max)} \mathbf{u}) / p_H^{(\max)} d$ . Upon variation of  $\theta$  this difference vanishes changing its sign when  $\mathbf{p}_{\parallel}^{(\max)}$  becomes perpendicular to  $\mathbf{u}$  which is the in-plane component of the electron inter-plane hopping vector. Due to this effect an experimental study of the difference dependence vs. angle  $\theta$  permits to determine the hopping vector  $\mathbf{h}$  and thus the orientation of the Fermi surface cylinder warping.

To establish a relation with the results of reference [6] one can use an easily proven identity:

$$\bar{v}_z(P_z) = \frac{\partial E / \partial p_z}{\partial S / \partial E} = - \frac{\partial S(P_z) / \partial P_z}{\partial S / \partial E} = - \frac{\partial S(P_z) / \partial P_z}{2\pi m}, \quad (13)$$

where  $S$  is the area enclosed by electron orbit,  $m$  the cyclotron mass. According to equation (3),  $\bar{v}_z$  may vanish if the nominator vanishes or the denominator goes to infinity. The latter case, considered in details in [8], takes place in the presence of self-crossing trajectories. We shall not consider it here. Due to smallness of  $\eta$ , approximately  $m = m_0 / \cos \varphi$ , where  $m_0$  is the cyclotron mass at  $\varphi = \pi/2$ , so, the denominator does not have singularities. On the other hand, it was shown in [6] that there exist critical angles  $\varphi_c$  where the maximum and minimum values of  $S$  as function of  $P_z$  nearly coincide, so that  $S(P_z)$  practically does not vary with  $P_z$ . It follows from equation (3) that  $\bar{v}_z(P_z)$  nearly vanishes for these angles. Of course, they are exactly the angles given by equation (12).

For more accurate formulation let us expand the periodic dependence  $S(P_z)$  in a Fourier series. At a generic angle  $\varphi$  the first garmonic dominates and further garmonics are minor corrections due to condition (2). At the critical angles  $\varphi_c$  the amplitude of the first garmonic exactly vanishes (see (11)) and  $\bar{v}_z$  is determined by the second garmonic of the  $S(P_z)$  function which is  $\eta$  times smaller than the first one. Beside, the hopping between the next nearest planes with the amplitude  $t'$  may also contribute to the second harmonic a term of the relative magnitude  $t'/t$  [8]. Hence, due to equation (6) the ratio of the saturation values of  $\rho_{zz}$  (7) at generic and critical angles is very small, being of the order of  $(C_1 t / \epsilon_F + C_2 t' / t)^2$ , where  $C_1$  and  $C_2$  are some coefficients.

As the amplitude of SdH oscillations is proportional to  $(\partial^2 S / \partial P_z^2)^{-1/2}$  [10], it should have maxima exactly at the same angles, where the resistivity does [6]. This statement is confirmed experimentally [1, 2].

### 3. Experimental results and discussion.

For the experiment, four  $\beta$ -(ET)<sub>2</sub>IBr<sub>2</sub> single crystals were taken with the residual resistance ratio  $R(300)/R(4.2) = 2000$  to 3000. Measurements of the resistance,  $R$ , perpendicular to the

highly conducting (a, b)-plane were carried out by means of a standard ac four-probe method with pasted contacts in magnetic field up to 15 T at the temperature  $T = 1.4$  K.

The samples were mounted into a cell which could be rotated in two planes with respect to the external magnetic field direction. This arrangement provided measurements of the resistance angular dependences for the field rotating in any plane perpendicular to the crystal (a, b)-plane. Thus, after cooling the sample, one could perform the whole set of magnetoresistance measurements avoiding repeated thermocycling harmful for the crystal quality.

The error in determination of the crystal orientation with respect to the field direction was less than  $1^\circ$  for the  $\varphi$  and did not exceed  $5^\circ$  for the  $\theta$ , where  $\varphi$  and  $\theta$  are the angles shown in the inset in figure 2.

In figure 2 four dependences of the magnetoresistance on the angle  $\varphi$  are exemplified for different angles  $\theta$ . Strong angular oscillations are observed for every plane of field rotation, being the most pronounced when  $\theta$  is close to  $90^\circ$ . This fact forces us to attribute them to the mechanism discussed in section 2 rather than to the appearance of self-crossing orbits on the Fermi surface.

The plots of  $N$  versus  $\cot \varphi_c$ , where  $\varphi_c$  and  $N$  are, respectively, the angle and the number of a local maximum in the  $R$  vs.  $\varphi$  dependence, clearly demonstrate the periodicity of the oscillations in  $\cot \varphi_c$  (figure 3):  $N = A^\pm(\theta) + B^\pm(\theta) \cot \varphi_c^\pm$ , where the signs  $\pm$  correspond to positive and negative values of  $\varphi_c$ , respectively. We note that the experimental data are fitted well by straight lines for every  $N$ , while the theory predicts the oscillations periodicity only for big  $N$  and sufficiently small  $\varphi_c$  when  $\cot \varphi_c \gg 1$  (12). For a given  $\theta$  the straight lines corresponding to positive and negative values of  $\varphi_c$  intersect the  $N$ -axis, generally, at different points, although their slopes coincide within  $\pm 5\%$ , i.e.  $\Delta A(\theta) \equiv A^+(\theta) - A^-(\theta) \neq 0$ ,  $B^+(\theta) \approx -B^-(\theta)$ . The cut-off difference,  $\Delta A$ , changes its sign in the vicinity of  $\theta \approx 70^\circ \pm 5^\circ$  and at angles far enough from this angle approaches to the value  $\pm 0.3$ . According to the considerations of section 2, it means that electron hopping takes place along a direction tilted away from the  $c^*$ -axis in the plane forming the angle approximately  $70^\circ + 90^\circ = 160^\circ$  with the  $a$ -axis. The direction of the in-plane component  $\mathbf{u}$  of the hopping vector  $\mathbf{h}$  is shown in figure 4 and is close to the  $-\mathbf{a}_{\parallel}^*$  direction, where  $\mathbf{a}_{\parallel}^*$  is the component of the inversed lattice vector  $\mathbf{a}^*$  perpendicular to the  $c^*$ . Using the cited above value of  $\Delta A$  and equation (12) one can evaluate an angle  $\alpha$  between the hopping vector  $\mathbf{h}$  and the  $c^*$  direction:  $\alpha \equiv \arctan(u/d) \approx 6^\circ \div 8^\circ$ ,  $u/d \approx 0.11$ .

Using equation (12) one can determine the form of the Fermi surface transverse cross-section as described in Sec. 2. The results are shown in figure 4. The Fermi surface cross-section area turns out to amount to  $\approx 3.9 \times 10^{15} \text{ cm}^{-2}$  that is close to the value, obtained from the SdH oscillations frequency  $S_{\text{osc}} \approx 3.7 \times 10^{15} \text{ cm}^{-2}$ , which corresponds to 55% of the Brillouin zone area  $S_{\text{BZ}}$  [1]. The form of the cross-section is in qualitative agreement with that calculated in [11].

It should be noted that the sharp peak at  $\varphi = 0^\circ$  in the  $R$  vs.  $\varphi$  dependences diminishes when the angle  $\theta$  approaches to  $\sim 60^\circ$ , and becomes undetectable in the angle interval  $\theta = 65^\circ$  to  $75^\circ$  (see Fig. 2). As mentioned above, in the same angle interval the cut-off difference in  $N$  vs.  $\cot \varphi_c$  dependencies is small and changes its sign. With further increasing the  $\theta$  the peak sharply restores. Special efforts have been undertaken to prove that the effect is not a consequence of a non-controlled misalignment of the sample.

At present, it is not clear why the peak disappears. Its existence seems to be associated with the appearance of the open orbits along the Fermi surface cylinder at  $\varphi = 0$ . So, it is natural to suppose that at  $\theta \approx 70^\circ \pm 5^\circ$  the contribution of the open orbits to the conductivity suddenly decreases. This might happen, for example, if the cylinders are connected by necks in the plane perpendicular to the direction  $\theta \approx 70^\circ$ , so that closed instead of open orbits are formed at  $\varphi = 0$ . The existence of such necks should not influence markedly the applicability

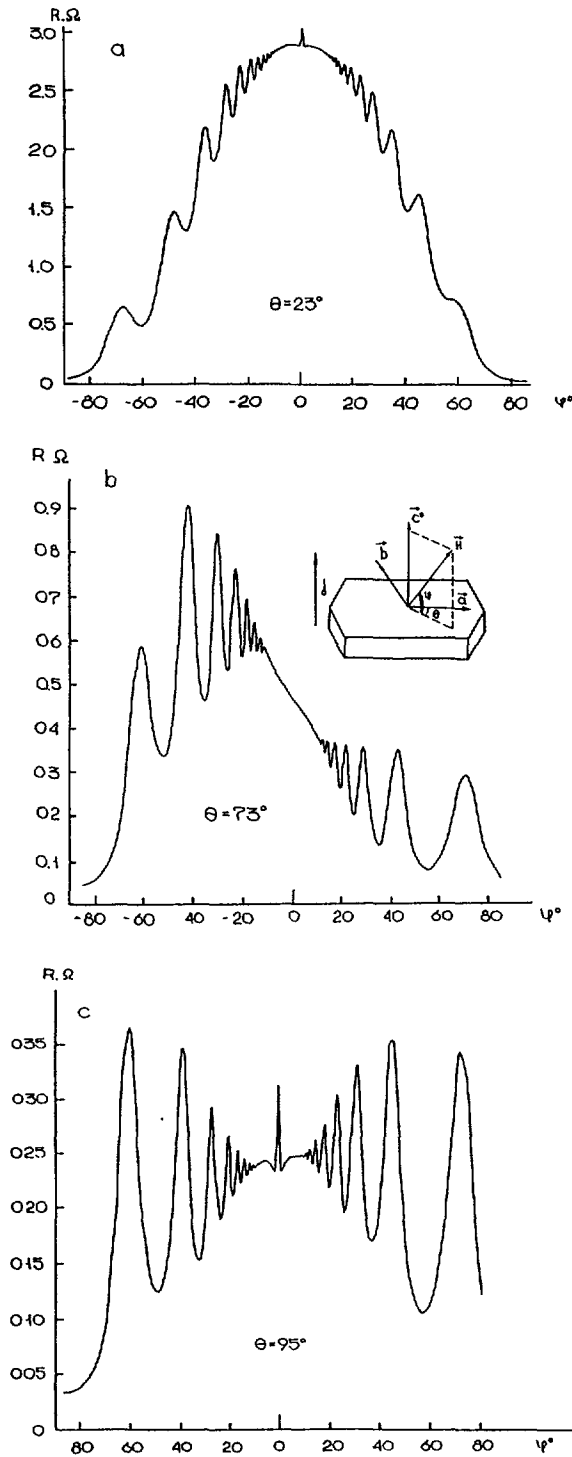


Fig. 2. — Examples of magnetoresistance dependences vs. angle  $\psi$  between the magnetic field direction and the (a, b)-plane for different angles  $\theta$ ; the angles  $\psi$  and  $\theta$  are shown in the inset.



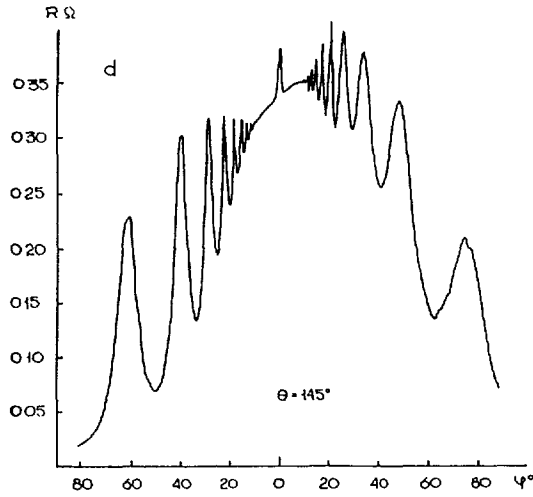


Fig. 2. — (continued)

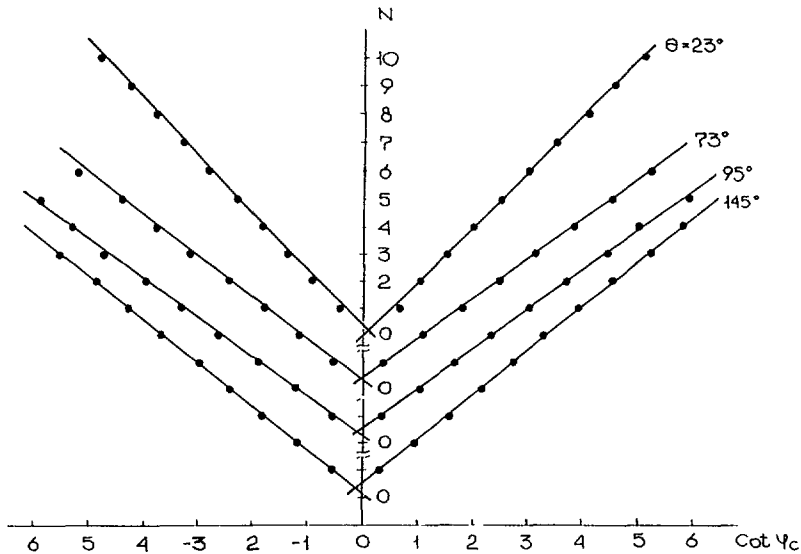


Fig. 3. — Dependences of the magnetoresistance local maximum number  $N$  on  $\cot \varphi_c$ , where  $\varphi_c$  are angles at which the local maxima are observed, for the field rotation planes presented in figure 2.

of the theoretical model considered in Section 2 provided the necks are sufficiently small and do not influence very much the Fourier expansion coefficients of the  $S(P_z)$  function at  $\varphi \neq 0$  (see equation (3)).

As it was noted in [12, 2, 1], the necks may be also responsible for slow SdH oscillations observed in crystals studied. The area corresponding to these oscillations is small,  $S \sim 5 \times 10^{13} \text{cm}^{-2} \approx 0.6\% S_{BZ}$ . The dependence of the oscillations frequency vs.  $\varphi$  at  $\theta = 0^\circ$  is shown in

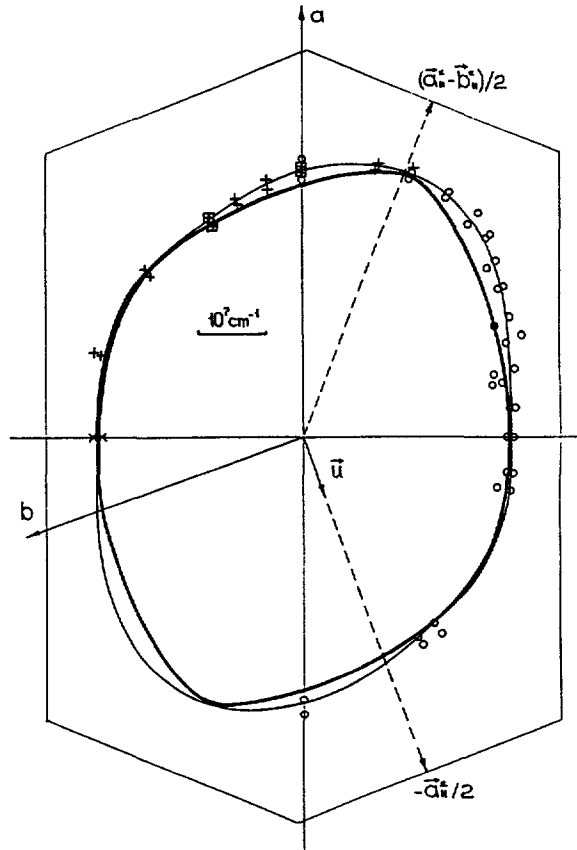


Fig. 4. — Transverse cross-section of the Fermi surface deduced from the  $p_H^{(\max)}(\theta)$  dependence as described in section 2 (thick line). Thin line interpolating experimental points represents the  $p_H^{(\max)}$  vs.  $\theta$  dependence. Different symbols denote four different samples. Vector  $u$  shows the direction of the in-plane component of the hopping vector. The magnitude of  $u$  is arbitrary. Framing lines indicate the Brillouin zone boundaries, the  $c^*$ -axis is directed upward with respect to the figure plane.

figure 5). It may be conjectured from this dependence that the area of an electron orbit sharply diminishes with tilting the magnetic field direction from the  $c^*$ -axis toward the  $-a$ -axis. This fact can be explained if the necks go along a direction tilted out of the  $(a, b)$ -plane. It is not excluded also that some pockets, lying between the Fermi surface cylinders and separated from them by a small gap, exist instead of the continuous necks. In this case a magnetic breakdown might occur at the particular field directions giving rise to a closed electron orbits.

In conclusion, we have studied the angular magnetoresistance oscillations in  $\beta$ -(ET)<sub>2</sub>IBr<sub>2</sub> under the magnetic field rotation in different planes perpendicular to the crystal  $(a, b)$ -plane. The existence of the oscillations for every rotation plane evidences that they are associated with quasi-two-dimensional character of the electron system, namely, with periodic vanishing of the mean electron velocity component along the magnetic field direction,  $v_H$ , in the case of a weakly corrugated cylindrical Fermi surface [6 – 9]. Using the obtained experimental data we have reconstructed the Fermi surface transverse cross-section of  $\beta$ -(ET)<sub>2</sub>IBr<sub>2</sub>. The analysis of the oscillations has provided an information about the electron interplane hopping vector. A

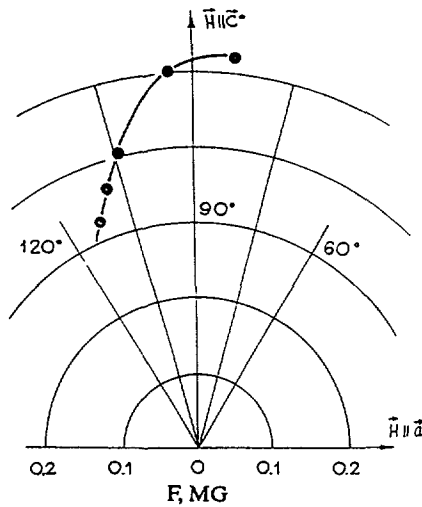


Fig. 5. — Dependence of the slow SdH oscillations frequency  $F$  vs.  $\varphi$  at  $\theta = 0^\circ$ .

sharp decrease of the contribution from open orbits to the in-plane magnetoresistance has been observed for the field direction perpendicular to the hopping vector. It might be associated with necks or pockets lying between the main Fermi surface cylinders.

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