Nonanalytical magnetoresistance, the third angular effect, and a method to investigate Fermi surfaces in quasi-two-dimensional conductors

A. G. Lebed
Electrotechnical Laboratory, 1-1-4 Umezono 350, Tsukuba, Japan;
Institute for Materials Research, Tohoku University, Sendai 980, Japan;
and L.D. Landau Institute for Theoretical Physics, 117334 Moscow, 2 Kosygina Street, Russia

N. N. Bagmet
Institute for Materials Research, Tohoku University, Sendai 980, Japan
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We demonstrate that transverse magnetoresistance is a nonanalytical function of the magnetic field, $\rho_\perp(H)\sim|H|^{1/2}$, if a magnetic field is parallel to the plane of anisotropy and normal to the Fermi surface at an inflection point in a quasi-two-dimensional (Q2D) conductor. The so-called ''third angular effect,'' recently discovered in organic conductors $(TMTSF)_2X$ $(X=ClO_4, PF_6)$ and $(DMET)_2I_3$, is interpreted in terms of the existence of an inflection point on their Fermi surfaces. Nonanalytical magnetoresistance is predicted to appear when the magnetic field is applied at the ''third magic angles,'' $\Theta=\pm\Theta_c$. It is also shown that at arbitrary directions of the in-plane magnetic field the magnetoresistance does not depend on relaxation time and obeys the law $\rho_\perp(H)\sim A|H|$ with factor A being a function of local characteristics of a Q2D Fermi surface. The above-mentioned phenomena provide useful methods to investigate Fermi surfaces in strongly anisotropic Q2D conductors including organic and high-T_c superconductors. [S0163-1829(97)51814-5]

Numerous quasi-two-dimensional (Q2D) conductors from chemical families $(TMTSF)_2X$ and $(ET)_2X$ demonstrate unusual properties in a metallic state in a magnetic field (for a review, see Refs. 1–4). Although such phenomena as ''magic angles'' and ''rapid magnetic oscillations'' seem to be of many-body origin,[3] some others have a clear Fermi-surface topology nature. Among them, there are Yamaji's,[5,4] Osada's,[6,4] Danner-Chaikin's,[7,1] and the so-called ''third''[8–10] angular resonances. Due to a strongly anisotropic nature of Fermi surfaces (FS's) in $(ET)_2X$ and $(TMTSF)_2X$ materials, the Fermi-surface topology effects in their metallic phases were shown to be nontrivial.[5–7,9,1,4]

The aim of our paper is to present nontrivial Fermi-surface topology phenomena that must exist in Q2D compounds. We point out that in the Q2D case (contrary to the 3D one[11]) the Boltzmann kinetic equation possesses solutions which diverge if the anisotropy and the value $\omega_c\tau$ tend to infinity. Under these conditions an electric current is defined by a small group of ''effective electrons'' which are located in the vicinity of the point P_H where the magnetic field is normal to the cross section of the Q2D FS (see Figs. 1 and 2). It is shown that it results in the appearance of non-analytical transverse magnetoresistance.

On the basis of the above-mentioned finding, we propose an interpretation of the recently discovered ''third angular effect'' (TAE)[8–10] and suggest useful methods for the investigation of Q2D FS's. Note that TAE was originally interpreted as a consequence of some changes in electron states.[8] In Ref. 9, this effect was treated in terms of the disappearance of closed orbits when the in-plane magnetic field is applied at the ''third magic angles'' (TMA's):

$$\Theta_c^{1,2}=\pm\arctan\left(\frac{2t_b b^*}{v_F}\right), \tag{1}$$

i.e., normal to the inflection point on the FS of a Q2D conductor with the electron spectrum:

$$\epsilon(\mathbf{p})=\pm v_F(p_a\mp p_F)-2t_b\cos(p_b b^*)-2t_\perp\cos(p_\perp c^*), \tag{2}$$

where $\epsilon_F=p_F v_F\simeq 2000$ K, $t_b\simeq 200$ K, and $t_\perp\simeq 5-10$ K; Θ is the angle between a axis and $\mathbf{H}$; $h=1$ (see Fig. 1).

Nevertheless, as shown in Ref. 10, TAE does not disappear if the magnetic field is slightly inclined with respect to the plane of anisotropy when closed orbits do not exist. Contrary to Ref. 9 (where numerical solutions of kinetic equation were found for the anisotropic 3D spectrum with an underestimated value of the anisotropy ratio, $t_b/t_\perp=4$), in the present paper we ignore the existence of small closed orbits. It is shown that TAE can be understood in terms of the appearance of nonanalytical magnetoresistance due to the existence of ''effective electrons'' in the case of large values of $t_b/t_\perp$ in Eq. (2). In addition, we show that, if the direction of the in-plane magnetic field is far enough from the TMA's

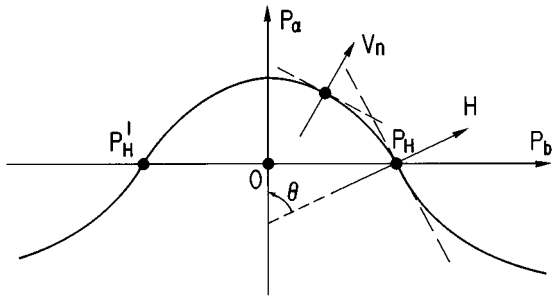

FIG. 1. The in-plane magnetic field is normal to the cross section, $p_a-p_F=2t_b\cos(p_b b^*)/v_F$, of the open FS (2) at the inflection point P_H.

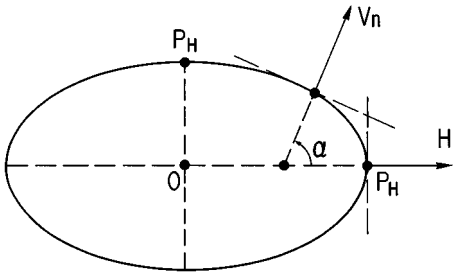

FIG. 2. The in-plane magnetic field is normal to the cross section, $\epsilon(p_a,p_b)=\epsilon_F$, of the Q2D FS (3) at the ordinary point P_H.

(1), magnetoresistance does not depend on electron relaxation time τ and depends only on some local characteristics of the FS. We propose experiments on the measurement of transverse magnetoresistance in Q2D compounds to obtain detailed information about their FS's.

Let us consider the peculiarities of the solutions of the kinetic equation in a strongly anisotropic Q2D conductor. For a conductor with an electron spectrum

$$\epsilon(\mathbf{p})=\epsilon(p_a,p_b)-2t_\perp\cos(p_\perp c^*),\quad t_\perp\ll\epsilon_F, \tag{3}$$

the Boltzmann kinetic equation

$$\left(eE+\frac{e}{c}[\mathbf{v}\times\mathbf{H}]\right)\frac{df(\mathbf{p})}{d\mathbf{p}}=-\frac{f(\mathbf{p})-f_0(\mathbf{p})}{\tau} \tag{4}$$

can be rewritten in the following form:

$$2eEt_\perp c^*\sin(p_\perp c^*)-\frac{e}{c}|\mathbf{v}_n(\alpha)|H\sin(\alpha)\frac{d\Psi(\mathbf{p})}{dp_\perp}=\frac{\Psi(\mathbf{p})}{\tau}\ , \tag{5}$$

if magnetic and electric fields are applied parallel and perpendicular to the plane of anisotropy. [In Eq. (5), we have omitted the component of the Lorentz force which comes from the electron velocity perpendicular to the plane; $f(\mathbf{p})=f_0(\mathbf{p})-[df_0(\mathbf{p})/d\mathbf{p}]\Psi(\mathbf{p})$; $\mathbf{v}_n$ is in-plane electron velocity; α is the angle between $\mathbf{H}$ and $\mathbf{v}_n$ (see Figs. 1 and 2)].

It is important that Eq. (5) has a simple solution,

$$\Psi(p_\perp,\alpha)=\frac{2eEt_\perp c^*\tau[\sin(p_\perp c^*)-\omega_c(\alpha)\tau\cos(p_\perp c^*)]}{1+\omega_c^2(\alpha)\tau^2}\ . \tag{6}$$

Therefore conductivity perpendicular to the (a,b) plane can be expressed in the following form:

$$\sigma_\perp(H)=\frac{e^2t_\perp^2c^*\tau}{\pi^2}\oint\frac{dl}{|\mathbf{v}_n(\alpha)|[1+\omega_c^2(\alpha)\tau^2]}, \tag{7}$$

where the integration is taken along the contour $\epsilon_\perp(p_a,p_b)=\epsilon_F$; $\omega_c(\alpha)=(e/c)|\mathbf{v}_n(\alpha)|Hc^*\sin\alpha$ is a characteristic frequency of an electron motion in the direction perpendicular to the plane. The main difference between Eqs. (6) and (7) and typical solutions of the kinetic equation in the 3D case[11,9] is that the integral (7) diverges at small α in the clean limit when $\omega_c\tau=\omega_c(90°)\tau\to\infty$.

Below, we consider two different cases: (1) 2D curvature R^{-1} of the cross section $\epsilon(p_a,p_b)=\epsilon_F$ of the FS's [(2) and (3)], is zero at point P_H (i.e., P_H is an inflection point) (see Fig. 1); (2) 2D curvature R^{-1} is nonzero at point P_H (i.e., P_H is an ordinary point) (see Fig. 2).

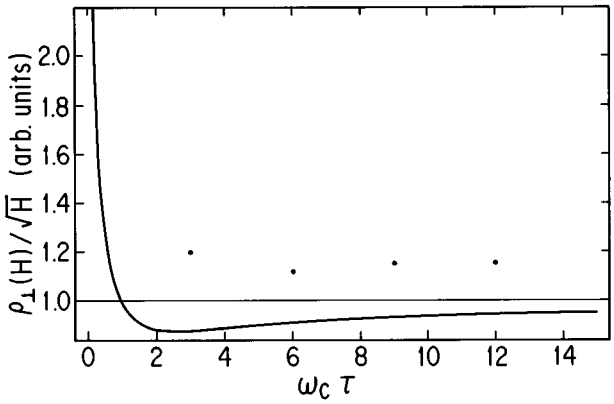

FIG. 3. The resistivity, calculated for the magnetic field applied at $\Theta=\Theta_c$ [Eq. (1)], is shown to obey a nonanalytical law, $\rho_\perp\sim|H|^{1/2}$, if $\omega_c\tau\geqslant 2$ (solid line). Solid circles: experimental data (Ref. 9) for $(TMTSF)_2ClO_4$.

It is easy to make sure that $\omega_c(\alpha)\sim\alpha\sim(dl)^2$ in the first case which results in a strong divergence of the integral (7) in the clean limit. From Eq. (7), it is possible to obtain the following expression for the perpendicular resistivity $\rho_\perp$ when $\omega_c\tau\gg 1$:

$$\rho_\perp(H)\ \sim\ \rho_\perp(0)\ (\omega_c\tau)^{1/2}\ \sim|H|^{1/2}. \tag{8}$$

[We have taken into account that $\sigma_\perp(H)\sigma_\parallel(H)\gg\sigma^2_{(\perp,\parallel)}(H)$ if $t_\perp\ll\epsilon_F$.]

We stress that magnetoresistance (8) is a nonanalytical function of a magnetic field. It is due to the fact that only ''effective electrons'' which are located in the vicinity of the inflection point P_H,

$$dl\sim\alpha^{1/2}\sim(\omega_c\tau)^{-1/2}\ll 1\ , \tag{9}$$

contribute to the conductivity (see Fig. 1). To demonstrate that nonanalytical behavior (8) is valid in a broad region of magnetic fields we performed a numerical estimation of the integral (7) for a model spectrum (2) of $(TMTSF)_2X$ compounds. Both the results of numerical calculations and experimental data[9] for $(TMTSF)_2ClO_4$ at ambient pressure are shown in Fig. 3. As it follows from Fig. 3, Eq. (8) (which has been derived in the limit $\omega_c\tau\gg 1$) becomes valid if $\omega_c\tau\geqslant 2$. From Fig. 3, it is also clear that magnetoresistance[9] measured at TMA is in satisfactory agreement with common nonanalytical dependence (8).

Let us consider the case when the in-plane magnetic field is applied at an arbitrary direction (i.e., P_H is not an inflection point) (see Fig. 2). In this case $\omega_c(\alpha)\sim\alpha\sim dl$ in Eq. (7) and the divergence of the integral (7) is not as strong as in the first case. Nevertheless, as follows from Eq. (7), transverse magnetoresistance is still a nonanalytical function of a magnetic field:

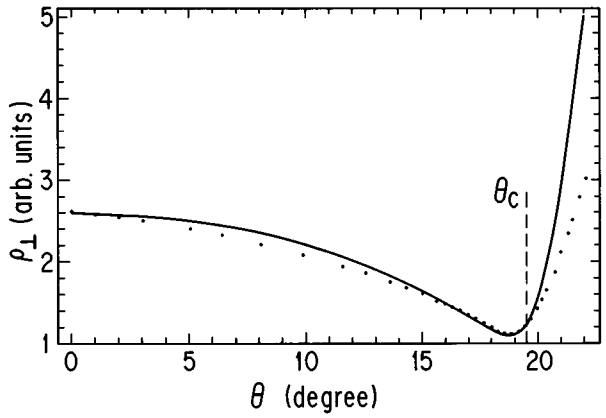

FIG. 4. Experimental data for $(TMTSF)_2PF_6$ (Refs. 10 and 12) (solid circles) are shown to be in a good agreement with theoretical angular dependence, $\rho_\perp(\Theta)$ (solid line), at $\Theta \leqslant \Theta_c \simeq 19.5°$.

$$\rho_\perp(H) \sim A(P_H) \ |H| \ , \qquad A(P_H) \sim \frac{|\mathbf{v}_n(P_H)|^2}{R(P_H)}. \quad (10)$$

Nonanalytical dependence (10) comes from the fact that for an arbitrary direction of the in-plane magnetic-field conductivity is defined by ''effective electrons'' located in the vicinity of the point P_H:

$$dl = R(P_H)\alpha \sim (\omega_c \tau)^{-1} \ll 1. \quad (11)$$

At this point we would like to present a qualitative explanation of TAE. When the direction of a magnetic field is approaching an inflection point P_H (see Fig. 1), more and more electrons are becoming ''effective'' [compare Eqs. (9) and (11)], which leads to the appearance of a minimum of the resistivity in the vicinity of the TMA's (1). To be more specific, we made numerical calculations of an angular dependence of the conductivity (7) for electron spectrum (2) with $t_a/t_b \simeq 8.3$ and compared it with the experimental angular dependence[10,12] that was obtained on the $(TMTSF)_2PF_6$ conductor (see Fig. 4).

Let us come back to Eq. (10). Note that magnetoresistance (10) does not depend on electron relaxation time τ and thus does not depend on temperature. This is an example of when the diagonal element of the resistivity tensor $\rho_\perp$ depends only on band-structure parameters. Unlike the nondiagonal Hall resistivity component $\rho(\perp,\|)$, transverse magnetoresistance (10) depends on local characteristics of a Q2D FS (3). Therefore, measurements of the angular dependence of the factor A in Eq. (10) gives information about the angular dependence of band parameter $|\mathbf{v}_n(P_H)|^2/R(P_H)$. In some cases the shapes of the FS's are known in Q2D compounds (see, for example, Ref. 13). Then measurement of an angular dependence of the magnetoresistance (10) provides a method to investigate the dependence of Fermi velocity, $|\mathbf{v}_n(P_H)|$, on the position on the FS. In a simple case of an elliptic Fermi surface we point out that $(OP_H)/(OP_H^*) = A(P_H)/A(P_H^*)$ (see Fig. 2). In the case of high-T_c compounds, when the existence of the FS's is not clear, the discovery of the magnetic-field dependence (10) could be a confirmation of an applicability of a Fermi-liquid picture.

Below, we discuss an applicability of the ''effective electrons'' approach (5)–(11) for the description of the properties of real Q2D compounds. The first question is ''When can we ignore the Lorentz force component which comes from the electron velocity perpendicular to the plane (i.e., ignore the existence of small closed orbits)?'' An analysis shows that for electron spectra (2) and (3) nonanalytical dependences (8) and (10) have to be valid in a broad region of magnetic fields, $1 \leqslant \omega_c \tau \leqslant (\epsilon_F/t_\perp)^{2/3}$ and $1 \leqslant \omega_c \tau \leqslant (\epsilon_F/t_\perp)^{1/2}$, correspondingly. Using the following values of the parameters $\epsilon_F \simeq 2000$ K, $t_\perp \simeq 5$ K, and $\tau \simeq 4.3 \times 10^{-12}$ sec[7], we found that for $(TMTSF)_2ClO_4$ the above inequalities can be rewritten as $1 \leqslant H \leqslant 40$ T and $1 \leqslant H \leqslant 15$ T. At higher and lower magnetic fields, solutions of the kinetic equation (4) are becoming nondivergent, which leads to the restoration of a textbook analytical magnetoresistance.[11,9]

The second important question is related to an applicability of the Fermi-liquid picture and kinetic equation (4) in the Q2D case. The comparison of a numerical estimation of the integral (7) with experimental data on $(TMTSF)_2PF_6$ (Refs. 10 and 12) at pressure $P = 8.5$ kbar (see Fig. 4) shows that the Fermi-liquid picture works well if there exist ''effective electrons'' on the FS (e.g., for the in-plane magnetic field applied at $\Theta < \Theta_c \simeq 19.5°$ [Eq. (1)]). The applicability of the kinetic equation at $\Theta \leqslant \Theta_c$ (which corresponds to $H_b \leqslant 2.3$ T) is also supported by the observation of angular oscillations in Refs. 10 and 12 which seem to be of a semiclassical origin.[10] On the other hand, in Refs. 14 and 15 it is shown that much smaller in-plane magnetic fields applied at $\Theta = 90°$, $H_b \simeq 0.2$ T, destroy coherent electron motion between layers in $(TMTSF)_2PF_6$ at pressure $P = 9.8$ kbar. Perhaps it indicates that pressure may induce a Fermi-liquid–non-Fermi-liquid transition in this compound.

In conclusion, we recall that above we ignore the existence of closed orbits in the case of in-plane (a-b) rotation of a magnetic field. The importance of closed orbits in $(TMTSF)_2ClO_4$ for out-of-plane (a-c) rotation was demonstrated both experimentally and theoretically in Ref. 7. Qualitative analysis shows that the ''effective electrons'' approach developed by us has to result in the appearance of a minimum of resistivity at the in-plane direction of a magnetic field under the conditions of out-of-plane rotation if $1 \leqslant \omega_c \tau \leqslant (\epsilon_F/t_\perp)^{1/2}$. This conclusion seems to be in accordance with the theoretical results of Ref. 7 where it is shown that the diminishing of $t_c = t_\perp$ results in the disappearance of a peak in resistivity at the in-plane direction of a magnetic field. We also consider the experimental observation of shallow minima at the in-plane direction of a magnetic field in $(TMTSF)_2ClO_4$ for $H \leqslant 6$ T (Ref. 7) as well as the observation of sharp minima at the in-plane direction of a magnetic field in $(ET)_2KHg(SCN)_4$ (see, for example, Ref. 17) as evidence of an importance of ''effective electrons'' in these compounds.

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