Angular Magnetoresistance Oscillations in Organic Conductors

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(Dated: November 15, 2004, Submitted to Phys. Rev. B, Rapid Commun.)

Abstract

We demonstrate that electron wave functions change their dimensionality at some commensurate directions of a magnetic field in conductors with open [quasi-one-dimensional (Q1D)] sheets of Fermi surface. These $1D \rightarrow 2D$ dimensional crossovers lead to delocalization of wave functions and are responsible for angular magnetoresistance oscillations. As an example, we show that suggested theory is in qualitative and quantitative agreements with the recent experimental data obtained on $(\text{TMTSF})_2\text{ClO}_4$ conductor.

PACS numbers: 74.70.Kn, 72.15.Gd
It is well known that, in traditional “three-dimensional” metals with closed quasi-particle orbits, quantum oscillations in a magnetic field occur due to Landau quantization of energy levels [1]. Meanwhile, a number of low-dimensional organic metals with open [quasi-one-dimensional(Q1D)] sheets of Fermi surfaces (FS) [2,3], where Landau quantization is not possible, demonstrate several types of unconventional angular oscillations [4-23] related to open sheets of FS,

\[
\epsilon^\pm(p) = \pm v_F \left( p_x \mp p_F \right) + 2t_b f(p_y b^*) + 2t_\perp \cos(p_\perp c^*) , \quad p_F v_F \gg t_b \gg t_\perp , \quad (1)
\]

where \(+(-)\) stands for the right (left) sheet of the FS; \(v_F\) and \(p_F\) are Fermi velocity and Fermi momentum along conducting \(x\)-axis, respectively; \(t_b\) and \(t_\perp\) are the overlapping integrals between conducting chains [2,3]; \(\hbar \equiv 1\).

Among these low-dimensional metals with Q1D sheets of FS (1), are \((\text{TMTSF})_2X\) (\(X = \text{PF}_6, \text{ClO}_4, \text{ReO}_4, \ldots\)) [8-19], \((\text{DMET})_2\text{I}_3\) [20], \(\kappa-(\text{ET})_2\text{Cu(NCS)}_2\) [21,22], \((\text{DMET-TTF})_2X\) (\(X = \text{AuCl}_2, \ldots\)) [23], and some other conductors. [For most Q1D conductors, one can use \(f(p_y b^*) = \cos(p_y b^*)\) in Eq.(1), with the important exceptions of \((\text{TMTSF})_2\text{ClO}_4\) and \((\text{TMTSF})_2\text{ReO}_4\) compounds [2,3]].

As was first recognized in theories of field-induced spin-density-wave phases in \((\text{TMTSF})_2X\) materials [24-27], in the absence of Landau quantization, Bragg reflections are responsible for quantum magnetic many-body phenomena in metals with open FS (1). Moreover, as shown in Ref.[4], at some Magic Angle (MA) directions of a magnetic field,

\[
\tan \alpha = N \left( b^*/c^* \right) , \quad \mathbf{H} = (0, H \sin \alpha, H \cos \alpha) , \quad (2)
\]
electron trajectories in a reciprocal space become periodic. This leads to the existence of constructive interference effects [28,29] coming from Bragg reflections, which occur when electrons move in a magnetic field along open Q1D sheets of FS (1) in the extended Brillouin zone. As a result, many-body angular oscillations in a magnetic field appear [4,5]. For extensions of the idea [4] to some model one-body phenomena, see Refs.[6,7].

Very recently, it has been theoretically demonstrated [28-31] that the similar interference effects are able to account for nontrivial one-body phenomena - angular magnetoresistance oscillations - experimentally observed in resistivity component \(\rho_{\perp}(\mathbf{H})\), perpendicular to conducting planes in \((\text{TMTSF})_2X\) [8-19] and \(\kappa-(\text{ET})_2\text{Cu(NCS)}_2\) [21,22] conductors. Note that the existence of angular magnetic oscillations [4-23] is an a contradiction with common belief that nothing important happens with open quasi-particles orbits (1) in a magnetic field and, as shown in Ref.[32], is a consequence of some non-trivial ”momentum quantization laws”.

In particular, in Refs.[28,29], a theory of angular magnetic oscillations in \(\rho_{\perp}(\mathbf{H})\) at MA directions of a magnetic field (2) was suggested, whereas, in Refs.[30,31], theoretical de-
scriptions of the so-called interference commensurate (IC) oscillations in \( \rho_\perp(\mathbf{H}) \) [19,20,33-35] were proposed. The IC magnetoresistance oscillations were also studied by numerical methods [19,33,34]. As shown in Refs.[29,28], the physical meaning of MA oscillations (2) in \( \rho_\perp(H, \alpha) \) is related to the interference effects between velocity component perpendicular to the conducting \((x,y)\)-planes, \( v_\perp(p_\perp) = -2t_\perp e^* \sin(p_\perp c^*), \) and the density of states. The IC oscillations are characterized by strong enough projection of a magnetic field on the conducting \( x \)-axis [19,20,33,34]:

\[
\mathbf{H} = H(\cos \theta \cos \phi, + \cos \theta \sin \phi, \sin \theta).
\]

(3)

This changes the physical meaning of the interference effects (see Ref.[30]), although minima in \( \rho_\perp(H, \theta, \phi) \) appear exactly at MA projection [19,30,31] of the field (3) on \((y,z)\)-plane, corresponding to the following commensurate angles [30,31]:

\[
\sin \phi = n(b^*/c^*) \tan \theta,
\]

(4)

where \( n \) is an integer.

Indeed, as shown in Ref.[30], interference effects, responsible for the IC oscillations, occur even for constant value of density of states [i.e., for \( v_F = \text{const} \) in Eq.(1)] and correspond to summation of infinite number of electron waves coming from some ”stationary phase points” [30] on electron trajectories in the extended Brillouin zone. Note that all existing theories of IC oscillations are developed for simplest Q1D spectrum with \( f(p_y b^*) = \sin(p_y b^*) \) in Eq.(1) and, thus, are not applicable to such typical Q1D conductor as \((\text{TMTSF})_2\text{ClO}_4\), where

\[
f(p_y b^*) = \sqrt{\cos^2(p_y b^*) + \Delta^2 / 4t_b^2},
\]

(5)

with \( \Delta \) being the so-called anion gap [2,3].

The main goals of our paper are as follows: 1) to calculate wave functions of a Q1D metal (1) with arbitrary function \( f(z) \) in an inclined magnetic field (3), 2) to reveal novel phenomenon - a change of space dimensionality of these wave functions from 1D to 2D at commensurate directions of a magnetic field (4), 3) to calculate \( \rho_\perp(H, \theta, \phi) \) and to show that it exhibits minima at commensurate directions (4) due to the above mentioned 1D \( \rightarrow \) 2D delocalizations of wave functions, 4) to demonstrate that these 1D \( \rightarrow \) 2D delocalizations manifest themselfs as saturations of magnetoresistance \( \rho_\perp(H, \theta, \phi) \), 5) to show that the suggested theory is in good agreement with the very recent experimental data obtained on \((\text{TMTSF})_2\text{ClO}_4\) [35].

Let us discuss how 1D \( \rightarrow \) 2D delocalization crossovers can result in the appearance of minima in \( \rho_\perp(H, \theta, \phi) \) using qualitative arguments. For electrons localized on conducting \( x \)-chains [4], resistivity component \( \rho_\perp(H, \theta, \phi) \) is expected to be infinite in the absence of impurities (i.e., at \( 1/\tau = 0 \)) \([1,7,28]\). At \( 1/\tau \neq 0 \), it has to demonstrate quadratic non-saturated
magnetoresistance, $\rho_{\perp}(H, \theta, \phi) \sim H^2 \tau$, in accordance with general theory [1,7] in the case of open quasi-particles orbits (1). If, at commensurate directions of the field (4), electrons become delocalized, then $\rho_{\perp}(H, \theta, \phi)$ is expected to have similarities with resistivity of a free electron at $H = 0$. Therefore, in this case, $\rho_{\perp}(H, \theta, \phi)$ has to saturate at high magnetic fields with the saturation values being expected to be proportional to $1/\tau$. Below, we demonstrate that this qualitatively different behavior of $\rho_{\perp}(H, \theta, \phi)$ at commensurate directions (4) is indeed responsible for the appearance of minima in $\rho_{\perp}(H, \theta, \phi) = 1/\sigma_{\perp}(H, \theta, \phi)$.

To develop an analytical theory, we make use of the Peierls substitutions method for an open electron spectrum [24,4,30]: $p_x - p_F \rightarrow -i(d/dx)$, $p \rightarrow p - (e/c)A$, $y \rightarrow i(d/dp_y)$. We choose the following vector potential for inclined magnetic field (3): $A = (0, x \sin \theta, x \cos \theta - y \cos \theta \cos \phi)H$, where Hamiltonian (1) in the vicinity of $p_x \simeq p_F$ can be expressed as

$$
\hat{e}^+(p) = -iv_F \left(\frac{d}{dx}\right) + 2t_b f \left[ p_y b^* - \frac{\omega_b(\theta)x}{v_F} \right] + 2t_{\perp} \cos \left[ p_{\perp} c^* + \frac{\omega_c(\theta, \phi)x}{v_F} \right] + i \left[ \hat{\omega}_c(\theta, \phi) \left(\frac{d}{dp_y}\right) \right],$$

$$
\omega_b(\theta) = eHv_F b^* \sin \theta/c, \omega_c(\theta, \phi) = \frac{eHv_F c^* \cos \theta \sin \phi}{c}, \hat{\omega}_c(\theta, \phi) = \frac{eHv_F c^* \cos \theta \cos \phi}{c}. \quad (6)
$$

It is possible to prove that, if one represents electron wave functions in the form

$$
\Psi_\epsilon(x, p_y, p_{\perp}) = \exp(ip_F x)\Psi^+_\epsilon(x, p_y, p_{\perp}), \quad (7)
$$

then solutions of the Schrodinger equation for Hamiltonian (6) can be written as

$$
\Psi^+_\epsilon(x, p_y, p_{\perp}) = \exp \left( \frac{iex}{v_F} \right) \exp \left( -\frac{2it_b}{v_F} \int_0^x f \left[ p_y b^* - \frac{\omega_b(\theta)u}{v_F} \right] du \right)
\times \exp \left( -\frac{2it_{\perp}}{v_F} \int_0^x \cos \left[ p_{\perp} c^* + \frac{\omega_c(\theta, \phi)u}{v_F} \right] + a \left( f \left[ p_y - \frac{\omega_b(\theta)u}{v_F} \right] - f[p_y] \right) \right) du \right),
\quad (8)
$$

Below, we demonstrate that suggested in the paper $1D \rightarrow 2D$ dimensional crossovers directly follow from Eq.(9). For this purpose, we calculate the real space $z$-dependence of wave functions along the inter-plane direction (i.e., at $z = Nc^*$, where $N$ is an integer plane index) by taking a Fourier transform of the second exponential function in Eq.(9):

$$
\Phi^+(x, p_y, z = Nc^*) = \int_0^{2\pi} \frac{dp_{\perp}}{2\pi} \exp(ip_{\perp}Nc^*)
\times \exp \left( -\frac{2it_{\perp}}{v_F} \int_0^x \cos \left[ p_{\perp} c^* + \frac{\omega_c(\theta, \phi)u}{v_F} \right] + a \left( f \left[ p_y b^* - \frac{\omega_b(\theta)u}{v_F} \right] - f[p_y b^*] \right) \right) du \right). \quad (9)
$$

After straightforward calculations, Eq. (10) can be expressed as

$$
\Phi^+(x, p_y, z = Nc^*) = \exp[-i\beta N] J_N \left[ \frac{2t_{\perp}}{v_F} \sqrt{R_1^2(x, p_y) + R_2^2(x, p_y)} \right], \quad (10)
$$
\[ I_{1,2}(x = \frac{2\pi M_0 v_F}{\omega_b(\theta)}, p_y) = \frac{1}{2^M} \sum_{M=0}^{M-1} \int_0^{2\pi f} \left( \exp \left[ \frac{\omega_c(\theta, \phi) u}{v_F} \right] + 2\pi M \frac{\omega_c(\theta, \phi)}{\omega_b(\theta)} \right) \]
\[ + a \left( f[p_y b^* - \frac{\omega_b(\theta) u}{v_F}] - f[p_y b^*] \right) \pm \text{c.c.} \, du, \]
\tag{11}
\]

with \( J_N(...) \) being the Bessel function \[36\]; \( M_0 \) is an integer, \( \beta \) is a phase factor. According to the theory of Bessel functions \[36\], \( J_N(Z) \) is an oscillatory function of variable \( N \) at \( N < |Z| \), whereas it decays exponentially with \( N \) at \( N > |Z| \). Thus, one can conclude that wave functions (11)-(13) are extended along \( z \)-direction if at least one of the functions \( I_i(...) \) in Eqs.(12),(13) is not restricted [i.e., if \( |I_i(M_0, p_y)| \rightarrow \infty \) as \( M_0 \rightarrow \infty \)]. In the opposite case, where both functions \( I_i(...) \) \((i = 1, 2)\) in Eqs.(12),(13) are not restricted \( |I_i(M_0, p_y)| < I_{max} \), wave functions (11)-(13) exponentially decay with the variable \( z \) at \( |z| = Nc^* \) \( \geq 4t_{\perp}^\perp I_{\max} / v_F \).

Note that functions (11)-(13) are written in the form of summations of infinite number of electron waves corresponding to quasi-classical electron motion in different Brillouin zones. Therefore, the physical meaning of summations in Eqs.(12),(13) is related to the interference effects due to Bragg reflections, which occur when electrons move along open orbits (1) in a magnetic field (3) in the extended Brillouin zone. As it is seen from Eqs.(12),(13), angular dependent phase difference between electron waves, \( 2\pi M \omega_c(\theta, \alpha)/\omega_b(\theta) \), is an integer multiple of \( 2\pi \) only at commensurate directions of a magnetic field (4), where \( \omega_c(\theta, \alpha) = n\omega_b(\alpha) \), with \( n \) being an integer. Therefore, one can conclude that, at arbitrary directions of the field (3), the destructive interference effects in Eq.(11) result in exponential decay of wave functions (9),(10) perpendicular to conducting \( x \)-chains, whereas, at commensurate directions (4), the constructive interference effects de-localizes electron wave functions (9),(10).

To calculate conductivity \( \sigma_{\perp}(H, \theta, \phi) \) within Fermi liquid approach for non-interacting quasi-particles (1), we introduce quasi-classical operator of the velocity component \( v_\perp \) in a magnetic field [30]:
\[ \hat{v}_\perp(p_{\perp}, x) = -2t_{\perp} c^* \sin \left[ p_{\perp} c^* + \frac{\omega_c(\theta, \phi) x}{v_F} \right] - i \left[ \frac{\hat{\omega}_c(\theta, \phi)}{v_F} \right] \left( \frac{d}{dp_y} \right) \]
\tag{12}
\]
and make use of Kubo formalism [7]. As a result, we obtain
\[ \sigma_{\perp}(H, \theta, \phi) \sim \int_0^{2\pi} dy \cos(z) \int_{-\infty}^{0} dz \exp(z) \int_{0}^{2\pi} \frac{dy}{2\pi} \left( \frac{\cos \phi}{\tan \theta} \right) (f[\omega_b(\theta) \tau z + y] - f[y]) \]
\tag{13}
\]
Eq.(15) is the main result of our paper. Since in Q1D \( \rho_{\perp}(H, \theta, \phi) = 1/\sigma_{\perp}(H, \theta, \phi) \), Eq.(15) solves a problem to determine resistivity \( \rho_{\perp}(H, \theta, \phi) \) for Q1D electron spectrum (1) with an arbitrary function \( f(p_y b^*) \) [37]. Note that the existence of commensurate minima
(4) in \( \rho_{\perp}(H, \theta, \phi) \) directly follows from Eq.(15). Indeed, the discussed above constructive interference effects correspond to commensurability of two frequencies in Eq.(15), \( \omega_c(\theta, \phi) = n\omega_b(\theta) \), where \( n \) is an integer. Thus, integral (15) increases at commensurate directions of the field (4) which leads to the appearance of minima in \( \rho_{\perp}(H, \theta, \phi) \).

It is possible to make sure that Eq.(15) predicts non-saturating magnetoresistance at high magnetic fields for the case, where wave functions (11)-(13) are localized (see Fig.1). This is in accordance with general theory of magnetoresistance for open quasi-particle orbits [1,7]. Here, we show that suggested in the paper 1D \( \rightarrow \) 2D dimensional de-localization crossovers manifest themselves as saturations of magneto-resistance \( \rho_{\perp}(H, \theta, \phi) \) at commensurate directions (4) of the field (3). For this purpose, we calculate \( \rho_{\perp}(H, \theta, \phi) = 1/\sigma_{\perp}(H, \theta, \phi) \) as a function of \( H \) for commensurate angle (4) with \( N = 1 \) for the most common case, where \( f(p_yb^*) = \cos(p_yb^*) \) in Eq.(1) [38]. As it is seen from Fig.1, de-localized electrons are characterized by unusual for open orbits (1) saturated behavior of magnetoresistance, \( \rho_{\perp}(H, \theta, \phi) \to \text{const} \) as \( H \to \infty \). In Fig.2, we present a comparison of our theory with the very recent measurements of \( \rho_{\perp}(H, \theta, \phi) \) performed on \((\text{TMTSF})_2\text{ClO}_4\) [35,38]. Fig. 2 demonstrates that our Eq.(15) is not only in qualitative but also in good quantitative agreement with the experimental results [35,38].

This work was supported in part by National Science Foundation, grant number DMR-0308973, the Department of Energy, grant number DoE-FG02-02ER63404, and by the INTAS grants numbers 2001-2212 and 2001-0791. One of us (AGL) is thankful to E.V. Brusse for useful discussions.

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From general equation (15), one can obtain its important limiting case for $f(p_y b^*) = \cos(p_y b^*)$ considered in Refs.\cite{30,31}.

For more detailed comparison of our Eq.\,(15) with recent experimental data obtained on (TMTSF)$_2$ClO$_4$, see Ref.\cite{35}.
\[ \rho_{\perp}(h) \text{(arb. units)} \]

FIG. 1: Magnetoresistance $\rho_{\perp}(H, \theta, \phi)$ as a function of "dimensionless magnetic field", $h = \omega_b(H, \theta = \pi/2) \tau$ [see Eq.(7)], calculated by means of Eq.(15) for Q1D electron spectrum (1) with $f(p_y b^*) = \cos(p_y b^*)$, $t_a/t_b = 8.5$, $b^* = c^*/2$. Upper curve: non-saturated magnetoresistance for localized electron wave functions (11)-(13) at $\theta = 3.5^\circ$ and $\phi = 2.6^\circ$. Lower curve: saturated magnetoresistance for de-localized wave functions (11)-(13) at $\theta = 3.5^\circ$ and $\phi = 1.75^\circ$, corresponding to commensurate direction (4) of a magnetic field (3) with $n = 1$. 


FIG. 2: Solid curve: magnetoresistance $\rho_\perp(H, \theta, \phi)$ as a function of angle $\phi$ calculated at $\theta = 7^\circ$ by means of Eq.(15) for $f(p_b b^*)$ given by Eq.(5) with $\Delta/2t_b = 0.1$, $t_a/t_b = 10$, $b^* = e^*/2$, $h = \omega_b(H, \theta = \pi/2)\tau = 15$. Dotes: experimental points [35] obtained on (TMTSF)$_2$ClO$_4$ conductor at $\theta = 7^\circ$ and $H = 10$ T [35,38].