Magic Angle Effects and Angular Magnetoresistance Oscillations as Dimensional Crossovers

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Interference effects between velocity and density of states, which occur as electrons move along open orbits in the extended Brillouin zone in anisotropic conductors, result in a change of wave functions' dimensionality at magic angle (MA) directions of a magnetic field. In particular, these 1D $\rightarrow$ 2D dimensional crossovers result in the appearance of sharp minima in a resistivity component $\rho_\perp(H, \alpha)$, perpendicular to conducting layers. This explains the main qualitative features of MA and angular magnetoresistance oscillations' phenomena observed due to the existence of quasi-one-dimensional sheets of Fermi surface in $(TMTSF)_2X$, $(DMET\text{-}TSeF)_2X$, and $\kappa\text{-}(ET)_2Cu(NCS)_2$ conductors.

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Low-dimensional organic conductors such as $(TMTSF)_2X$ ($X = PF_6, ClO_4, \ldots$), $(DMET\text{-}TSeF)_2X$ ($X = AuCl_2, \ldots$), and $\kappa\text{-}(ET)_2Cu(NCS)_2$ exhibit a number of unconventional angular magnetic oscillations [1–24] related to quasi-one-dimensional (Q1D), open Fermi surface (FS) sheets [1–3].

Such systems can be described by a dispersion relation,

$$\epsilon^{\pm}(\mathbf{p}) = \pm v_F(p_x \mp p_F) - 2t_b \cos(p_y b^*) - 2t_\perp \cos(p_\perp c^*), \quad p_F v_F \gg t_b \gg t_\perp, \tag{1}$$

where + (−) stands for the right (left) sheet of the FS; v_F and p_F are the Fermi velocity and Fermi momentum along the most conducting $\mathbf{x}$ axis, respectively; t_b and $t_\perp$ are wave function overlap integrals along the $\mathbf{y}$ and $\mathbf{z}$ directions (between the conducting chains); $\hbar \equiv 1$. Most unconventional angular oscillations in a metallic phase—the so-called Danner-Kang-Chaikin oscillations [17], the third angular effect [18–20], and the interference commensurate oscillations [20,21]—have been explained in terms of a Fermi liquid (FL) approach to anisotropic Q1D spectrum (1) (see Ref. [17], Ref. [25], and Refs. [26,27], correspondingly).

On the other hand, despite the fact that all experimentally observed "magic angle" (MA) phenomena [5–16] and angular magnetoresistance oscillation (AMRO) [22–24] are related to MA directions [4,28,29] of a magnetic field, via

$$\tan\alpha = (n/m)(b^*/c^*), \qquad \mathbf{H} = (0, H\sin\alpha, H\cos\alpha) \tag{2}$$

(where n and m are integers) corresponding to periodic electron orbits in the $(p_y, p_\perp)$ plane [4,29], there is as yet incomplete agreement between the numerous theories of MA phenomena [28–39] and experiments [5–16] in the metallic phase. There even exists experimental evidence that, although some MA effects in the metallic phase [7,16] are of FL origin, others [3,12–14] may significantly deviate from a FL picture.

The goal of our Letter is to demonstrate that electron wave functions, corresponding to an open FS in a realistic tight-binding model of a Q1D spectrum, change their dimensionality from 1D to 2D at MA directions of a magnetic field (2) with $m = 1$:

$$\tan\alpha = n(b^*/c^*). \tag{3}$$

This spectrum contains electron hopping only between neighboring molecular sites, with

$$\epsilon^{\pm}(\mathbf{p}) = \pm v_x(p_y)[p_x \mp p_x(p_y)] - 2t_\perp \cos(p_\perp c^*), \quad p_x(p_y) = p_F + 2t_b \cos(p_y b^*)/v_F. \tag{4}$$

In particular, we show that, in the absence of Landau level quantization for open FS (4), the other quantum effects in a magnetic field, Bragg reflections, result in 1D $\rightarrow$ 2D dimensional crossovers at MA directions (3).

In other words, electron wave functions, which are localized on the conducting *chains* at arbitrary directions of a magnetic field [4,40], become 2D (i.e., localized on *planes*) at the MA directions of the field (3). As shown below, the nontrivial physical origin of these 1D $\rightarrow$ 2D dimensional crossovers is related to interference effects between the velocity component $v_\perp(p_\perp)$, perpendicular to the conducting planes, and the density of states, which is proportional to $1/v_x$, as electrons move along open FS (4) in the extended Brillouin zone. Using this finding, we demonstrate that it is possible to explain the appearance of MA [5,7,13,15,16] and AMRO [22–24] minima in the resistivity component $\rho_\perp(H, \alpha)$, perpendicular to the conducting planes in $(TMTSF)_2X$, $(DMET\text{-}TSeF)_2X$, and $\kappa\text{-}(ET)_2Cu(NCS)_2$ compounds, in the framework of a FL approach.

At first, let us discuss how 1D $\rightarrow$ 2D dimensional crossovers lead to the appearance of MA minima in $\rho_\perp(H, \alpha)$ using qualitative arguments. For electrons localized on conducting $\mathbf{x}$ chains [4,40], it is natural to expect [31,41] that the conductivity component $\sigma_\perp(H, \alpha)$ is zero in the absence of impurities (i.e., at $1/\tau = 0$) and decays as

$1/\tau H^2$ at high fields in the presence of impurities [31]. If, at MA directions of the field (3), electron wave functions become delocalized, then $\sigma_\perp(H, \alpha)$ is expected to have similarities with the conductivity of a free electron at $H = 0$. Therefore, in this case, $\sigma_\perp(H, \alpha)$ has to saturate at high magnetic field and is expected to be proportional to τ. Below, we demonstrate that this qualitatively different behavior of $\sigma_\perp(H, \alpha)$ at MA directions (3) is indeed responsible for the appearance of MA minima in $\rho_\perp(H, \alpha)$.

To develop a quantitative theory, we make use of the Peierls substitution method [41] for an open orbit electron spectrum [4,42]: $p_x \to -i(d/dx)$, $p_y \to p_y - (e/c)A_y$, $p_\perp \to p_\perp - (e/c)A_z$. It is convenient to choose a vector potential in the form $\mathbf{A} = (0, Hx\cos\alpha, -Hx\sin\alpha)$, where Hamiltonian (4) in the vicinity of $p_x \simeq p_F$ can be expressed as

$$\hat{\epsilon}^{+}(\mathbf{p}) = v_x\left[p_y b^* - \frac{\omega_b(\alpha)x}{v_F}\right]\left(-i\frac{d}{dx} - p_x\left[p_y b^* - \frac{\omega_b(\alpha)x}{v_F}\right]\right) - 2t_\perp \cos\left[p_\perp c^* + \frac{\omega_c(\alpha)x}{v_F}\right] \tag{5}$$

with

$$\omega_b(\alpha) = eHv_F b^* \cos\alpha/c, \qquad \omega_c(\alpha) = eHv_F c^* \sin\alpha/c \tag{6}$$

being cyclotron frequencies of electron motion perpendicular to the conducting chains. An important difference between Hamiltonian (5) and the Hamiltonians [4,27,42] studied so far is that the velocity component along the conducting $\mathbf{x}$ chains, related to an operator of the density of states, $1/\hat{v}_x$, depends on p_y and x. While in this case, operators $\hat{v}_x$ and d/dx in Eq. (5) do not commute, one can nevertheless ignore this fact if the quasiclassical parameter

$$4t_\perp/\omega_b(\alpha) \gg 1. \tag{7}$$

If one represents the electron wave functions in the form

$$\Psi_\epsilon(x, p_y, p_\perp) = \exp\left(i\int_0^x p_x\left[p_y b^* - \frac{\omega_b(\alpha)u}{v_F}\right]du\right)\psi_\epsilon(x, p_y, p_\perp), \tag{8}$$

then the solutions of the Schrödinger equation for Hamiltonian (5) can be written as

$$\psi_\epsilon(x, p_y, p_\perp) = \frac{1}{\sqrt{v_x[p_y b^* - \frac{\omega_b(\alpha)x}{v_F}]}}\exp\left(i\int_0^x \frac{\epsilon du}{v_x[p_y b^* - \frac{\omega_b(\alpha)u}{v_F}]}\right)\exp\left(2it_\perp\int_0^x \frac{\cos[p_\perp c^* + \frac{\omega_c(\alpha)u}{v_F}]}{v_x[p_y b^* - \frac{\omega_b(\alpha)u}{v_F}]}du\right). \tag{9}$$

[In Eq. (9), we have normalized the wave functions by the standard condition, $\int \psi_{\epsilon_1}(x)\psi_{\epsilon_2}(x)dx = \delta(\epsilon_1 - \epsilon_2)$, and made use of inequality (7)].

Let us demonstrate that 1D $\to$ 2D dimensional crossovers directly follow from Eq. (9). It is possible to prove that in the limiting case, where $v_x = v_F = \text{const}$, wave functions (8) and (9) are *always* localized on conducting chains (see Refs. [4,40]). Below, we show that accounting for dependences of the density of states on p_y and x, $1/v_x[p_y b^* - \omega_b(\alpha)x/v_F]$ in Eq. (9), leads to delocalization crossovers at MA directions of the field (3). For this purpose, we calculate the real-space dependence of the electron wave functions along the interplane direction (i.e., at $z = Nc^*$, where N is an integer plane index) by taking the Fourier transform of the second exponential function in Eq. (9):

$$\Phi(x, p_y, z = Nc^*) = \int_0^{2\pi}\frac{dp_\perp}{2\pi}\exp(ip_\perp Nc^*)\exp\left(2it_\perp\int_0^x \frac{\cos[p_\perp c^* + \frac{\omega_c(\alpha)u}{v_F}]}{v_x[p_y b^* - \frac{\omega_b(\alpha)u}{v_F}]}du\right). \tag{10}$$

After straightforward calculations, this z dependence of electron wave functions (10) can be expressed as

$$\Phi(x, p_y, z = Nc^*) = \exp[-i\phi N]J_{-N}[2t_\perp\sqrt{I_1^2(x, p_y) + I_2^2(x, p_y)}], \tag{11}$$

where

$$\begin{aligned} I_1(x = 2\pi M_0 v_F/\omega_b(\alpha), p_y) &= \sum_{M=0}^{M_0}\int_0^{2\pi v_F/\omega_b(\alpha)} \frac{\cos[\frac{\omega_c(\alpha)u}{v_F} + 2\pi M\frac{\omega_c(\alpha)}{\omega_b(\alpha)}]}{v_x[p_y b^* - \omega_b(\alpha)u/v_F]}du, \\ I_2(x = 2\pi M_0 v_F/\omega_b(\alpha), p_y) &= \sum_{M=0}^{M_0}\int_0^{2\pi v_F/\omega_b(\alpha)} \frac{\sin[\frac{\omega_c(\alpha)u}{v_F} + 2\pi M\frac{\omega_c(\alpha)}{\omega_b(\alpha)}]}{v_x[p_y b^* - \omega_b(\alpha)u/v_F]}du, \end{aligned} \tag{12}$$

with J_N being the Bessel function [43], M_0 an integer, and ϕ a phase factor. According to Bessel function theory [43], $J_N(Z)$ is an oscillatory function of the variable N for $N < |Z|$, whereas it decays exponentially with N for $N > |Z|$. Thus, one can conclude that wave functions (10)–(12) are extended along the $\mathbf{z}$ direction if at least one of the functions I in

Eq. (12) is not restricted [i.e., if $|I_i(M_0, p_y)| \to \infty$ as $M_0 \to \infty$]. In the opposite case, where both functions I_1 and I_2 are restricted by the conditions $|I_{1,2}(M_0, p_y)| < I_{\max}$, electron wave functions (10)–(12) decay exponentially with the variable z for $|z = Nc^*| \geq 2I_{\max}$.

Note that functions (12) are written in the form of summations of an infinite number of electron waves, corresponding to quasiclassical electron motion in different Brillouin zones in an extended zone picture. Therefore, the physical meaning of the summations in Eq. (12) is related to the interference effects between velocity component $v_\perp = -2t_\perp c^* \sin[p_\perp c^* + \omega_c(\alpha)u/v_F]$ and the density of states, $1/v_x[p_y b^* - \omega_b(\alpha)u/v_F]$, which occur due to Bragg reflections in a magnetic field. As is seen from Eq. (12), the angular dependent phase difference between electron waves, $2\pi M\omega_c(\alpha)/\omega_b(\alpha)$, is an integer multiple of 2π only at MA directions, where $\omega_c(\alpha) = n\omega_b(\alpha)$, with n an integer.

To calculate the conductivity $\sigma_\perp(H, \alpha)$ within a FL approach for noninteracting quasiparticles (4), let us introduce a quasiclassical operator of the velocity component $v_\perp$ in a magnetic field [27]:

$$\hat{v}_\perp(p_\perp, x) = -v_\perp^0 \sin[p_\perp c^* + \omega_c^0(\alpha)x/v_F], \qquad v_\perp^0 = 2t_\perp c^*. \tag{13}$$

Since wave functions (8)–(10) and the velocity operator (13) are known, one can calculate $\sigma_\perp(H, \alpha)$ by means of Kubo formalism. As a result, one obtains

$$\sigma_\perp(H, \alpha) \sim \left\langle \frac{1}{v_x(p_y)} \int_{-\infty}^{0} d(b^*u) \frac{\cos[n(\alpha)b^*u]}{\omega_b(p_y + u, \alpha)} \times \exp\left[- \int_u^0 \frac{d(b^*u_1)}{\tau\omega_b(p_y + u_1, \alpha)} \right] \right\rangle_{p_y}, \tag{14}$$

where

$$\omega_b(p_y, \alpha) = \omega_b(\alpha)[v_x(p_y)/v_F], \quad \omega_c(p_y, \alpha) = \omega_c(\alpha)[v_x(p_y)/v_F], \quad n(\alpha) = \omega_c(\alpha)/\omega_b(\alpha), \tag{15}$$

and $\langle \cdots \rangle_{p_y}$ stands for averaging over p_y.

After straightforward but rather complicated integrations, Eq. (14) can be rewritten as

$$\frac{\sigma_\perp(H, \alpha)}{\sigma_\perp(0)} = \frac{1}{1 + h_c^2(H)} - h_c^2(H) \int_{-\infty}^{0} du \exp(u) \cos[h_c(H)u] \left\langle \exp\left[\int_0^u f(y + u_1 h_b(H)) du_1 \right] - 1 \right\rangle_y, \tag{16}$$

$$f(y) = v_F/v_x(y) - 1, \qquad h_b(H) = \omega_b(\alpha)\tau, \qquad h_c(H) = \omega_c(\alpha)\tau.$$

Since in Q1D, $\rho_\perp(H, \alpha) \simeq 1/\sigma_\perp(H, \alpha)$, Eq. (16) solves the problem of defining $\rho_\perp(H, \alpha)$ for electrons with open orbit spectrum (4) in inclined magnetic field (2).

To make our results more intuitive, we consider an important limiting case of Eq. (16)—a so-called clean limit, where $\omega_c(\alpha)\tau \gg 1$. In this case, Eq. (16) can be significantly simplified:

$$\frac{\sigma_\perp(H, \alpha)}{\sigma_\perp(0)} = \frac{1}{1 + [\omega_c(\alpha)\tau]^2} - \tan^2\alpha \left(\frac{c^*}{2b^*} \right)^2 \sum_{n=1}^{\infty} \frac{A_n^2}{n^2} \left(\frac{2}{1 + [\omega_c(\alpha)\tau]^2} - \frac{1}{1 + [\omega_c(\alpha) - n\omega_b(\alpha)]^2\tau^2} - \frac{1}{1 + [\omega_c(\alpha) + n\omega_b(\alpha)]^2\tau^2} \right), \tag{17}$$

where A_n are the Fourier coefficients of function $f(y) = v_F/v_x(y) - 1$:

$$A_N = \frac{1}{\pi} \int_{-\pi}^{+\pi} f(y) \cos(Ny) dy. \tag{18}$$

Equation (16) [and its clean limit (17)] is the main result of this Letter, and is distinct from all other models of conduction in such anisotropic systems. Equation (17) directly demonstrates that $\sigma_\perp(H, \alpha)$ maxima [$\rho_\perp(H, \alpha)$ minima] are related to minima in the denominators which occur at MA defined by $\omega_c(\alpha) = n\omega_b(\alpha)$ or Eq. (3). In Fig. 1, we present numerical simulations of Eqs. (17) and (18) for three qualitatively different variants of Q1D spectrum (4), corresponding to $(TMTSF)_2PF_6$, κ-$(ET)_2Cu(NCS)_2$, and $(TMTSF)_2ClO_4$ conductors. As is seen, $(TMTSF)_2PF_6$ exhibits only one MA minimum, while the others exhibit several minima with large indices n in Eq. (3). We stress that all these qualitative features, as well as a doubling of the period of MA minima in the case of $(TMTSF)_2ClO_4$, are in good agreement with existing experimental data [5,7,13,16,23] and can be related to peculiarities of the Q1D electron spectra in the above compounds (see figure caption).

We point out that the existing alternative model to describe MA and AMRO effects in $\rho_\perp(H, \alpha)$, due to Osada *et al.* [31], while important from methodological and historical points of view, lacks direct physical meaning. The reason is that the transfer integrals $t_{n,m}$ in that model are exponentially small in the framework of a realistic tight-binding model [1] of the Q1D electron spectrum. The 1D $\to$ 2D dimensional crossovers and interference effects suggested in this Letter have a real physical origin, related to the p_y dependence of the density of states, $1/v_x(p_y)$, in Eqs. (4) and (5). From a classical point of view as first pointed out by Maki [32], in our case, the interference effects occur because of the fact that electrons spend a disproportionate amount of time on those parts of FS (4) where the density of states is larger. Such effects are not possible within the Osada model [31] and all its variants, which use a linearized electron spectrum

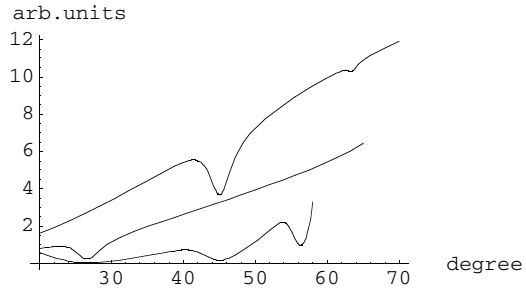

FIG. 1. Resistivity component $\rho_\perp(\alpha) = 1/\sigma_\perp(\alpha)$, perpendicular to the conducting layers, calculated by means of Eqs. (17), (18), and (4). For κ-$(ET)_2Cu(NCS)_2$ (lower curve) and $(TMTSF)_2PF_6$ (middle curve), we use Q1D electron spectrum $\epsilon(p_x, p_y, p_\perp) = 2t_a \cos(p_x a/2) + 2t_b \cos(p_y b^*) + 2t_\perp \times \cos(p_\perp c^*)$ [1] with weak, $t_a/t_b \simeq 3$, and strong, $t_a/t_b = 8.5$ [27], Q1D anisotropies [1], respectively. For $(TMTSF)_2ClO_4$ (upper curve), we take into account anion ordering [1] and thus use the spectrum $\epsilon(p_x, p_y, p_\perp) = 2t_a \cos(p_x a/2) + \sqrt{[2t_b \cos(p_y b)]^2 + \Delta^2} + 2t_\perp \cos(p_\perp c)$ [1] with $t_a/t_b = 8.5$ and $\Delta = 0.2t_b$. In all three cases, we utilize the values $\omega_b(0)\tau = 15$ and $c^* = 2b^*$. Note that for layered conductors (i.e., at $t_\perp \ll t_b$) our theoretical results do not depend on the precise value of $t_\perp/t_b$.

with a *constant* density of states. For example, the weighting factors in Eq. (17) depend on magnetic field orientation (i.e., on $\tan\alpha$) and, thus, their physical meaning is completely different from the angle-independent "effective transfer integrals" postulated in Ref. [31].

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