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Quantum Theory of Lee–Naughton–Lebed’s Angular Effect in Strong Electric Fields

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Abstract: Some time ago, Kobayashi et al. experimentally studied the so-called Lee–Naughton–Lebed’s (LNL) angular effect in strong electric fields [Kobayashi, K.; Saito, M.; Omichi E.; Osada, T. Phys. Rev. Lett. 2006, 96, 126601]. They found that strong electric fields split the LNL conductivity maxima in an a-(ET)₂-based organic conductor and hypothetically introduced the corresponding equation for conductivity. In this paper, for the first time, we suggest the quantum mechanical theory of the LNL angular oscillations in moderately strong electric fields. In particular, we demonstrate that the approximate theoretical formula obtained by us well describes the above mentioned experiments.

Keywords: quantum mechanics; quasi-two-dimensional conductor; high magnetic field

1. Introduction

It is well known that organic conductors having quasi-one-dimensional (Q1D) pieces of the Fermi surfaces (FSs) demonstrate unique magnetic properties due to the Bragg reflections of moving electrons from the Brillouin zone boundaries in moderate and strong magnetic fields [1–5]. Among them are the Field-Induced Spin(Charge)-Density-Wave (FIS(C)DW) phase diagrams [3–15], 3D Quantum Hall Effect (3D QHE) [14–16], the so-called Lebed’s Magic Angles (LMAs) [17–40], the Lee–Naughton–Lebed’s (LNL) angular oscillations [41–47], and some others. Note the LMA phenomena [17–40] seem to be very complicated and, in most cases, possess some non-Fermi liquid (FL) properties [1,27,29], whereas the FIS(C)DW, 3D QHE, and LNL phenomena have been successfully explained in the framework of the Landau FL approach [1,2]. In particular, the LNL phenomenon has been successfully theoretically explained in Refs. [48–54]. Indeed, in Refs. [48–54], a layered Q1D conductor with the electron spectrum was considered,

\[ \varepsilon_{\pm}^0(p) = \pm v_F(p_x \mp p_F) + 2t_b \cos(p_y b^*/\hbar) + 2t_c \cos(p_z c^*/\hbar). \]  

[Note that in Equation (1), the first term represents electron free motion along the conducting chains on the right (+) and left (−) sheets of the Q1D FS, with \( p_F \) and \( v_F \) being the Fermi momentum and Fermi velocity, correspondingly. The second and the third term correspond to the hopping of electrons in the perpendicular axes, \( b^* \) and \( c^* \) (\( p_F v_F \gg t_b \gg t_c \)); \( p \) is the total electron momentum; \( p_x \) is its component along conducting chains, whereas \( p_y \) and \( p_z \) are electron momentum components along \( b^* \) and \( c^* \) axes, correspondingly; \( \hbar \) is the Planck constant.] The Q1D conductor is placed in the following inclined magnetic field,

\[ H = H \sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta, \]  

and in electric field \( E \) along \( z \) direction (see Figure 1). In the quasi-classical approximation, the following expression for the LNL conductivity was derived by several methods:

\[ \sigma_{zz}(H, \theta, \phi) = \sigma_{zz}(0) \sum_{N=-\infty}^{\infty} \frac{\int_{N}^\infty \left[ \omega_c(\theta, \phi)/\omega_b(\theta) \right]}{1 + \int_{N}^\infty \left[ \omega_c(\theta, \phi) - N\omega_b(\theta) \right]^2 K}. \]  

where $\sigma_{zz}(0)$ is conductivity at $H = 0$, and $J_N(x)$ is the Bessel function of the $N$-th order. Note that in Equation (3), the so-called electron cyclotron frequencies can be expressed as [48–54]:

$$\omega_b(\theta) = \frac{|e|v_F H b^* \cos \theta}{\hbar}, \quad \omega_c(\theta, \phi) = \frac{|e|v_F H c^* \sin \theta \sin \phi}{\hbar},$$

$$\omega_c^*(\theta, \phi) = \frac{|e|v_0^* H c^* \sin \theta \cos \phi}{\hbar}, \quad v_0^* = 2t_b b^*, \quad v_y = 2t_b b^*,$$

where $e$ is the electron charge, and $c$ is the speed of light. More recently, Kobayashi et al. [55] experimentally studied the LNL phenomenon in rather strong electric fields and found that the strong electric field splits the LNL maxima of conductivity (3). What is also important is that they suggested a hypothetical theoretical formula which described the above mentioned experimental splitting.

![Figure 1. Definition of the azimuthal angle $\theta$ and polar angle $\phi$ for the typical Lee–Naughton–Lebed’s experiment, where $z$ is the least conducting axis.](image)

The goal of our paper is to derive the quasi-classical expression for conductivity in moderately strong electric and strong magnetic fields which describes the experimentally observed splitting of the LNL maxima of conductivity [55]. In particular, we show that our equation has a limited area of applicability and is not applicable in very strong electric fields.

2. Materials and Methods

First, let us perform the quasi-classical Peierls substitution [56,57] for motion along the conducting chains in Equation (1), in the absence of both magnetic and electric fields.

$$\hat{\epsilon}_0^\pm(x, p_y, p_z) = \mp i\frac{\hbar}{2t_b} \frac{d}{dx} + 2t_b \cos(p_y b^*/\hbar) + 2t_c \cos(p_z c^*/\hbar).$$

The solution of the corresponding Schrödinger equation is

$$\Psi_0^\pm(x, p_y, p_z) = \exp \left( \mp i\frac{\epsilon x}{v_F \hbar} \right) \exp \left[ \mp i\frac{2t_b x}{v_F \hbar} \cos(p_y b^*) \right] \exp \left[ \mp i\frac{2t_c x}{v_F \hbar} \cos(p_z c^*) \right],$$

where energy $\epsilon$ is counted from the Fermi level, $\epsilon_F = p_F v_F$. 

Then, we introduce the electric field applied along the least conducting \( z \) axis as a small perturbation to the Hamiltonian (6),

\[
\delta \hat{e}(z) = eEz,
\]

and perform one more quasi-classical Peierls substitution [56,57]:

\[
\delta \hat{e}(p_z) = eEz = -ie\hbar \frac{d}{dp_z}.
\]

In this case, the application of the perturbation (9) to the free electron wave function (7) gives

\[
\delta \Psi(\rho) = \pm \frac{eEx}{v_F\hbar} 2t_e c^+ \sin(p_c c^+ / \hbar) \Psi(\rho).
\]

It is easy to prove that, for not extremely strong electric fields, the total Hamiltonian in the electric field can be written as

\[
\hat{\mathcal{H}}(x, y, p_z) = \mp i\hbar v_F \frac{d}{dx} + 2t_b \cos(p_y b^+ / \hbar) + 2t_c \cos \left( \frac{p_c c^+}{\hbar} + \frac{eEz c^+}{v_F} \right).
\]

Here, we introduce the magnetic field (2) in the electron Hamiltonian and the electron velocity operator along \( z \) axis. For further development, it is convenient to choose the vector potential of the magnetic field in the following form:

\[
\mathbf{A} = (0, x \cos \theta, -x \sin \theta \sin \phi + y \sin \theta \cos \phi) \mathbf{H}.
\]

To define the corresponding electron wave functions for the case, where \( t_b \gg t_c \), as shown in Ref. [51], it is necessary to take into account only two first terms in Hamiltonian (11) and to perform in the second term the following quasi-classical Peierls substitution,

\[
p_y \rightarrow p_y - \frac{\mathcal{A}_{y}}{c}.
\]

In this case, wave function in the mixed \((x, p_y)\) representation obeys the following Schrödinger equation [3,51]:

\[
\left( \mp i\hbar v_F \frac{d}{dx} + 2t_b \cos(p_y b^+ / \hbar) - \frac{\omega_b(\theta)x}{v_F} \right) \Phi^\pm(x, p_y) = e\Phi^\pm(x, p_y),
\]

where the two wave functions (7) and (14) are related by the following equation:

\[
\Psi^\pm(x, p_y) = \exp(\pm ip_F x / \hbar) \Phi^\pm(x, p_y).
\]

It is important that Equation (14) can be exactly solved,

\[
\Phi^\pm(x, p_y) = \exp \left( \pm \frac{i}{v_F} \frac{p_c c^+}{\hbar} \right) \exp \left( \pm \frac{2it_b}{\hbar v_F} \frac{\omega_b(\theta)x}{v_F} - \frac{p_y b^+}{\hbar} \right) \left( \sin \left[ \frac{p_y b^+}{\hbar} - \frac{\omega_b(\theta)x}{v_F} \right] - \sin \left[ \frac{p_y b^+}{\hbar} \right] \right). \tag{16}
\]

Let us apply the quasi-classical Peierls substitution to energy dependence (11) to the momentum component along \( z \) axis:

\[
\hat{\mathcal{H}}^\pm(x, y, p_z) = 2t_c \cos \left( \frac{p_z c^+}{\hbar} + \frac{eEz c^+}{v_F} \right) + 2t_c \cos \left[ \frac{p_z c^+}{\hbar} + \frac{eEz c^+}{v_F} + \frac{\omega_c(\theta, \phi)x}{v_F} + \frac{\omega_c^+(\theta, \phi)y}{v_F} \right].
\]

Taking into account that, in the quasi-classical approximation,

\[
\delta^\pm(x, y, p_z) = \frac{d[\mathcal{H}^\pm(x, y, p_z)]}{dp_z}, \quad y = -ib(d/dp_y), \tag{18}
\]
it is possible to write the velocity component operator along \( z \) axis in the following form:

\[
\sigma_z^\pm (x, y, p_z) = -2lt_e c^+ \sin \left[ \frac{p_z c^+}{\hbar} \mp eEc^+x \frac{v_F}{v_F h} + \frac{\omega_c^+ (\theta, \phi) x}{v_F} - i \frac{\omega_c^+ (\theta, \phi) h (d/dp_y)}{v_y^0} \right]. \tag{19}
\]

In Equation (19), for further development, we introduce

\[
\omega_c^\pm (\theta, \phi) = \omega_c (\theta, \phi) \mp eEc^+ / h. \tag{20}
\]

It is important that wave functions (16) are eigenfunctions of the velocity operator along \( z \) axis (19), (20) with the following eigenvalues:

\[
\Phi^\pm_e (x, p_y) = -2lt_e c^+ \sin \left[ \frac{p_y b^+}{\hbar} \pm \omega_b (\theta) \right] \times \left( \cos \left[ \frac{p_y b^+}{\hbar} - \frac{\omega_b (\theta) x}{v_F} \right] - \cos \left[ \frac{p_y b^+}{\hbar} \right] \right) \Phi_e (x, p_y). \tag{21}
\]

3. Results

Let us make use of the Kubo formula for conductivity \([51, 58]\). We can do this because the electron wave functions (16) and the eigenvalues of velocity operators (21) are known. The total conductivity along \( z \) axis can be represented as a summation of the following two contributions: one from the right sheet of the FS (1) and another from the left sheet,

\[
\sigma_{zz} (H, \theta, \phi) = \sigma_{zz}^+ (H, \theta, \phi) + \sigma_{zz}^- (H, \theta, \phi). \tag{22}
\]

By means of the Kubo formalism \([51, 58]\), we obtain

\[
\sigma_{zz}^\pm (H, \theta, \phi) \sim \int_{-\pi}^{\pi} \frac{d(p_y b^+)}{2} \int_0^\infty \frac{dx}{v_F} \exp \left( - \frac{x}{v_F \tau} \right) \cos \left[ \frac{\omega_c^+ (\theta, \phi) x}{v_F} \pm \frac{\omega_c^+ (\theta, \phi)}{\omega_b (\theta)} \left( \cos \left[ \frac{p_y b^+}{h} - \frac{\omega_b (\theta) x}{v_F} \right] - \cos \left[ \frac{p_y b^+}{h} \right] \right) \right]. \tag{23}
\]

where \( \tau \) is an electron relaxation time. The complicated double integration in Equation (23) can be simplified using definitions of the Bessel functions of the N-th order, \( f_N (x) \) \([51, 59]\),

\[
\sigma_{zz}^\pm (H, \theta, \phi) = \frac{\sigma_{zz} (0)}{2} \sum_{N = -\infty}^{\infty} \frac{f_N^2 [\omega_c^+ (\theta, \phi) / \omega_b (\theta)]}{1 + \tau^2 [\omega_c^+ (\theta, \phi) - N \omega_b (\theta)]^2}, \tag{24}
\]

where \( \sigma_{zz} (0) \)—conductivity along \( z \) axis in low electric fields in the absence of the magnetic field. If we make use of Equation (22), we finally obtain for the total conductivity in moderately strong electric fields in the presence of the inclined magnetic field (2) the following:

\[
\sigma_{zz} (H, \theta, \phi) = \frac{\sigma_{zz} (0)}{2} \sum_{N = -\infty}^{\infty} \left\{ \frac{f_N^2 [\omega_c^+ (\theta, \phi) / \omega_b (\theta)]}{1 + \tau^2 [\omega_c^+ (\theta, \phi) - N \omega_b (\theta)]^2} + \frac{f_N^2 [\omega_c^+ (\theta, \phi) / \omega_b (\theta)]}{1 + \tau^2 [\omega_c^+ (\theta, \phi) - N \omega_b (\theta)]^2} \right\}. \tag{25}
\]

4. Discussion

We stress that Equation (25) is the main result of our paper, whereas in Ref. \([55]\), this equation was just guessed. Moreover, we have shown that it is not exact and has to be used for not too high (i.e., moderately high) electric fields. Indeed, let us discuss its applicability. We recall that we have derived Equation (25) using some approximation: we have suggested that we can use Equation (11), instead of Equation (10). It is easy to prove that this can be done under the condition that

\[
\frac{|e| Ec^+ x_0}{v_F h} \ll 1, \tag{26}
\]
where $x_0$ is characteristic length where the integral (23) converges. Since, as follows from (23), $x_0 \simeq v_F \tau$, the condition (26) can be written as

$$|e|Ec^* \ll \hbar / \tau.$$  \hspace{1cm} (27)

If we take the lowest experimentally used electric field, $V_0 = Ed = 2 \text{ V}$, $d = 0.2 \text{ mm}$ [55], $\hbar / \tau = 2 \text{ K}$ and $c^* \simeq 2 \text{ nm}$ [1], we obtain the inequality (27) in the form

$$0.25K \ll 2K,$$  \hspace{1cm} (28)

which shows that, at lowest voltages, the analysis in [55] is correct, whereas at higher experimental voltages like $V_0 = 20 \text{ V}$ [55], Equation (25) must be used with great caution, since Equation (27) gives quantities of the same orders of magnitudes for the left side and for the right one.

Let us briefly discuss one important consequence of Equation (25)—the splitting of the LNL maxima of conductivity in moderately strong electric fields [55]. In the limit of zero electric field at the following typical experimental conditions, where

$$\omega_b(\theta) \gg 1, \quad \omega_c(\theta, \phi) \tau \gg 1,$$  \hspace{1cm} (29)

the maxima of conductivity, as follows from Equation (3), appear at

$$\omega_c(\theta, \phi) = N\omega_b(\theta),$$  \hspace{1cm} (30)

where $N$ is an arbitrary integer. Under the experimental condition (29), Equation (25) splits each maximum into two ones, which are defined by the following equations

$$\omega_c^{1,2}(\theta, \phi) = N\omega_b(\theta) \mp \omega_E, \quad \omega_E = eEc^* / \hbar.$$  \hspace{1cm} (31)

The effect of splitting was experimentally observed in Ref. [55]. Our analysis of the applicability of Equation (25), as we discussed above, has shown that Equation (25) is valid for lower experimentally used voltages, $V_0 \simeq 2 \text{ V}$, and become controversial at higher ones, $V_0 \simeq 20 \text{ V}$.

It is interesting that the obtained results are general for all families of the Q1D conductors. The splitting of the LNL maxima of conductivity appears due to the fact that the Lorentz force changes its sign between the left and right pieces of the Q1D FS due to the change in the electron velocity sign, whereas the electric force does not change its sign. Therefore, it is instructive to analyze novel Equations (26) and (27) in the typical type of Q1D conductors like the (TMTSF)$_2$X conductors. Indeed, the LNL oscillations are best studied in (TMTSF)$_2$PF$_6$, where $c^* = 1.36 \text{ nm}$ [1] and $\hbar / \tau \simeq 1 \text{ K}$ [43]. As seen from Equation (27), the splitting of the LNL maxima of conductivity has to be observed in the same electric field range as they were observed in the $\alpha$-(ET)$_2$-based conductor by Kobayashi et al. [55]. The obvious experimental problem is how to avoid the overheating of the (TMTSF)$_2$PF$_6$ sample. As for the (TMTSF)$_2$ClO$_4$ conductor, we have to be careful and use the Peierls substitution method at magnetic fields which are lower than the so-called magnetic breakdown field [1].

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