Effect of electronic band dispersion curvature on de Haas-van Alphen oscillations

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(Dated: August 5, 2015)

The effect of electronic band curvature, i.e. the deviation from parabolicity of electronic dispersion, on de Haas-van Alphen oscillations spectra is studied. Although the oscillations amplitude remain unaffected, it is demonstrated that non-quadratic terms of the Landau bands dispersion, which is particularly relevant in the case of Dirac fermions, induces a field- and temperature-dependent Onsager phase. As a result, a temperature-dependent shift of the de Haas-van Alphen oscillations frequency is predicted.

PACS numbers: 71.10.Ay, 71.18.+y, 73.22.Pr

Keywords: Magnetic oscillations, de Haas-van Alphen oscillations, Dirac fermions

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I. INTRODUCTION

Magnetic oscillations or de Haas-van Alphen effect (dHvA) in quasi-two-dimensional metals are well accounted for by the Lifshitz-Kosevich (LK) theory [1–5], which relates the frequency to the surface area enclosed by the cyclotronic trajectories. This geometrical approach is based on the semi-classical quantization theory of Onsager [6], and allows for the determination of many physical parameters of the Fermi surface (FS). Characteristic field \( B_c = \Phi_0/A_0 \) associated with the quantum flux trough the unit cell area \( A_0 \) is generally very large. Indeed, for organic conductors with unit cell area as large as \( A_0 = 100 \text{Å}^2 \), \( B_c \) is still 4136 T. Therefore, available magnetic fields stay well within the limit of the semi-classical approximation. In the opposite case, e.g. for significantly larger unit cell or applied magnetic fields, quantum corrections to the Landau spectrum [7] or modification of the Lifshitz-Kosevich (LK) theory would be necessary.

A question that remains to be addressed deals with the effect of departure from parabolic curvature of the electronic band dispersion on the amplitude and phase of quantum oscillations. This question is particularly relevant in the case of Dirac fermions, the electronic dispersion of which is linear. In this case, the LK calculation based on a truncation at first order in energy when evaluating the grand potential is no more sufficient, since the Landau level energy is known to display a square root dependence on the Landau level index [8].

In this paper, we address the question of the relevance of the non-parabolicity in two classes of materials. First, the LK calculation for magnetization in the presence of a uniform field is reconsidered in the case of Landau quasiparticles, or conventional fermions, relevant to e.g. organic conductors. Then Dirac fermions, which have linear band dispersion, are considered. In both cases the field- and temperature-dependent phase factor of the Fourier coefficients is evaluated as a function of the FS curvature which is the main parameter of the model. We can also mention other work dealing with non-parabolicity of the Fermi surface, in the special case of a tight-binding model in two dimensions [9] where the band gap closes for certain filling factor, and where the temperature amplitude does not follow the LK formula.

The main results of this paper can be summarized as follow. For a band dispersion in a two dimensional material with a closed Fermi surface, we assume that the area of such surface is given by the quantity \( S(E) \) at energy \( E \), close to the Fermi energy \( E \simeq \mu \). The
effect of band curvature (defined as the second derivative $S''(E) = \partial^2 S(E)/\partial E^2$ near the Fermi energy $E = \mu$) is mainly to add a phase shift contribution $\phi_p$ in each harmonics of order $p$ of the oscillating quantities such as the magnetization. More precisely, the phase shift is a function of field and temperature and is given by the following semi-classical expression in the case of organic conductors

$$\phi_p = \frac{\pi^2 k_B T h^2 S''(\mu)}{m^*} \varphi\left(2\pi^2 p \frac{k_B T}{\hbar \omega_c}\right),$$

$$\varphi(x) = \left[\sinh(2x) - x - x \cosh^2(x)\right]/\sinh^2(x), \quad (1)$$

where $m^*$ is the effective mass of the quasi-particle and $\omega_c = eB/m^*$ the cyclotronic frequency. In the case of Dirac fermions, for which the Landau energy level spectrum $E_n$ increases like the square root of the index $n$, the expression of the phase shift is given instead by

$$\phi_p = \frac{\pi}{2 m^* v_F} \varphi\left(2\pi^2 p \frac{k_B T}{\hbar \omega_c}\right),$$

$$\varphi(x) = \left[2 \sinh(2x) - x - x \cosh^2(x) - 2x^{-1} \sinh^2(x)\right]/\sinh^2(x). \quad (2)$$

where $v_F$ is the Fermi velocity. In the following we will derive both Eq. (1) and Eq. (2) using semi-classical analysis, and study their asymptotic limits, when both $T$ or $B$ are varied within physical ranges. This effect has to be differentiated from other possible contributions coming from additional physical mechanisms. For example, the presence of a spin-orbit coupling leads to a splitting of the dHvA frequency whose magnitude is proportional to $B^2$ and effective mass $m^*$ [10]. Frequency splitting due to spin-orbit coupling has been studied in bilayer high-$T_c$ cuprates [11] and can also be attributed to the splitting of the Fermi surface. Additional phases exist in the presence of magnetic breakdown. It is well known that magnetic breakdown is accompanied with a field-dependent phase [12], especially for large orbits. This phase depends more precisely on the ratio between the field and the breakdown field, and not temperature. Existence of this Onsager phase has been studied and revealed in organic compound $\theta$-(BEDT-TTF)$_4$CoBr$_4$(C$_6$H$_4$Cl$_2$) where the breakdown field is close to 35T [13]. More recently, the existence of phase offsets $\gamma$ has been questioned in thermodynamic quantities of three-dimensional topological insulators with surface states [14–16]. The topological nature of these insulators can be detected due to the presence of a Berry phase within the oscillations when the particle-hole symmetry is broken and the material has a band gap [17]. These global phases (as for the Maslov index $\gamma$, see below)
are independent of temperature, but present a linear variation with the field, contrary to the field and temperature-dependent phases Eq. (1) and Eq. (2) which come from a local effect of the band dispersion as we will see in the next section. Non-zero topological Berry’s phase was also investigated in graphene by measuring the magnetoresistance in the quantum Hall regime, with a compelling evidence of a value for $\gamma$ different from 1/2 due to the presence of Dirac fermions. For a review of the topological phases in condensed matter physics, see [19].

II. EFFECT OF BAND CURVATURE CORRECTIONS IN ORGANIC CONDUCTORS

Within the semi-classical framework, the phase quantization can be expressed in terms of surface swept by the quasiparticle in the Brillouin zone. It is given by the integral

$$S(E_n = E) = \frac{1}{4\pi^2} \oint_{E=E(k_x,k_y)} k_y dk_x = \pm b(n + \gamma),$$

where $b = eB/h$ is an effective Planck constant or reduced field, and $E_n$ is the energy of the Landau band with $n$ the Landau level index. $\gamma$ is the Maslov index which is equal to 1/2 for Landau quasiparticles with a parabolic band. In such a case, $S(E) = m^*E/(2\pi\hbar^2)$ varies linearly with the energy. For massless Dirac electrons with a linear energy dispersion, $S(E) = \pi E^2/(2\pi\hbar v_F)^2$ and $\gamma = 0$. In two-dimensional systems, the grand potential is expressed by

$$\Omega(B, \mu, T) = -A b \sum_{n=0}^{\infty} \log(1 + \exp[\beta(\mu - E_n)])$$

where $\mu$ is the chemical potential and $A$ is the sample area. $bA$ is the degeneracy of each Landau Level. The Poisson formula can be used for any discrete series over positive integers $n$: $\sum_{n=0}^{\infty} F(n) = \int_{0}^{\infty} F(n) \, d\eta + 2 \sum_{p=1}^{\infty} \int_{0}^{\infty} F(n) \cos(2\pi pn) \, d\eta$. This allows us to rewrite the oscillating part of the grand potential in terms of Fourier modes

$$\frac{\Omega_{osc}}{A} = -\frac{2b}{\beta} \text{Re} \sum_{p=1}^{\infty} \int_{0}^{\infty} \log(1 + \exp[\beta(\mu - E_n)]) \exp(2i\pi pn) \, d\eta.$$

Using $\exp(2i\pi pn) \to \exp(2i\pi pn)/(2i\pi p)$ as primitive function, a double integration by parts can be performed, yielding

$$\frac{\Omega_{osc}}{A} = -2b \text{Re} \sum_{p=1}^{\infty} \int_{0}^{\infty} \frac{\beta E_n^2}{4 \cosh^2 \left[ \frac{\beta}{2} (E_n - \mu) \right]} \frac{\exp(2i\pi pn)}{(2i\pi p)^2}.$$
\[-\frac{\beta E_n E_n''}{4 \cosh^2 \left[ \frac{\beta}{2} (E_n - \mu) \right]} \exp \left( \frac{2i \pi p n}{(2i \pi p)^3} \right) \], \quad \text{Eq. (5)}

where \( E'_n = \partial E_n / \partial n \) and \( E''_n = \partial^2 E_n / \partial n^2 \). Since for a parabolic band \( E''_n = 0 \), only the first term does not vanish in this case and \( E'_n = \hbar \omega_c \) is \( n \)-independent and proportional to \( b \). In general \( E''_n, E'''_n \), etc... are non zero, and Eq. (5) can be solved around \( E = \mu \) using the formal series expansion (see also [7])

\[ S(E) = S(\mu) + (E - \mu) \frac{\partial S}{\partial E}(\mu) + \frac{1}{2}(E - \mu)^2 \frac{\partial^2 S}{\partial E^2}(\mu) + \cdots \]  

In the standard LK theory, only the first two terms in Eq. (6) are taken into account: the first one typically sets the frequency of the oscillations, while the second one (linear in the energy difference) gives rise to the thermal reduction factor (see page 184 of reference [2] after equation 10.28 for instance). The third term studied in this manuscript, and quadratic in energy difference, is responsible for a phase shift of the oscillations as we will see further below. To illustrate the discussion, we can take a typical example, the tight binding model of free electrons on a discrete lattice with hopping parameter \( t \) and which is described by the energy dispersion \( E(k_x, k_y) = -t[\cos(k_x) + \cos(k_y)] \). The density of states can be written as

\[ A_0 S(E) = \frac{4}{\pi^2} \int_{-2}^{E/t} \frac{1}{2 - u} \left[ K \left( \frac{2 + u}{2 - u} \right) \right] u, \]  

where \( A_0 \) is the area of the unit cell and \( K(k) \) is the complete elliptic integral of the first kind: \( K(k) = \int_0^1 \frac{u[(1 - u^2)(1 - k^2 u^2)]^{-\frac{1}{2}}}{u} \) \( \text{Eq. (7)} \) is computed from Eq. (3) using the coordinate equation \( k_y = \cos^{-1}(-E/t - \cos(k_x)) \), then differentiating it with respect to the energy to get \( S'(E) \). After the change of variable \( u = \cos(k_x) \), Eq. (7) can be rewritten as

\[ A_0 \frac{\partial S(E)}{\partial E} = \frac{1}{\pi^2 t} \int_{-1-E/t}^{1} \frac{u}{\sqrt{(1-u^2)(1-(u+E/t)^2)}} \]  

After the additional change of variable \( v = u + \frac{1}{2}E/t \), the integral is symmetric around the origin, and a further transformation \( w = v/(1 + \frac{1}{2}E/t) \) leads to an expression involving the elliptic integral. We can in particular perform an expansion around the lower band limit \( E = -2t \) such that

\[ A_0 S(E) = \frac{1}{2\pi} \left( \frac{E}{t} + 2 \right) + \frac{1}{16\pi} \left( \frac{E}{t} + 2 \right)^2 + \cdots \]  

We can identify \( S'(E) = (2\pi A_0 t)^{-1} \) with \( 2\pi m^*/\hbar^2 \), since \( 4t = 4\hbar^2 / (A_0 m^*) \) is the bandwidth. The curvature parameter of the surface area enclosed by the orbit defined in the following
by $\kappa = S''(E)$, can be rewritten as $\kappa = (8\pi A_0 t^2)^{-1}$. Therefore for a given dispersion, we can relate the different coefficients of the expansion in Eq. (6) with microscopic parameters such as the hopping constants, effective mass or bandwidths.

In the most general case, we would like to use the expansion Eq. (6) up to second order to compute the different thermodynamic quantities such as Eq. (4). Up to now, the LK calculation considers only the first order around the Fermi surface, whose coefficient is given by the effective mass (the slope of $S(E)$). The second order term will modify, as we will see below, mostly the phase of the magnetic oscillations. To include the effect of the second order term, we perform first a change of variable $n \rightarrow E$ in the expression of the grand potential Eq. (5) using

$$\Omega_{osc} = -2b \Re \sum_{p=1}^{\infty} \int_{0}^{\infty} \left[ \frac{\beta E'_n}{4 \cosh^2 \left( \frac{\beta}{2} (E - \mu) \right)} \exp \left[ 2i\pi pn(E) \right] \right] E.$$

We then use the relations

$$E'_n = \frac{\hbar \omega_c}{1 + \tilde{\kappa}(E_n - \mu)}, \quad E''_n = -\frac{\tilde{\kappa}(\hbar \omega_c)^2}{1 + \tilde{\kappa}(E_n - \mu)^2}; \quad \tilde{\kappa} = \frac{2\pi \hbar^2 \kappa}{m^*},$$

(10)
to obtain the oscillating part of the grand potential expressed as an integral over the energy

$$\frac{\Omega_{osc}}{A} = -2b \Re \sum_{p=1}^{\infty} \int_{0}^{\infty} \left[ \frac{\beta \hbar \omega_c}{4[1 + \tilde{\kappa}(E_n - \mu)] \cosh^2 \left( \frac{\beta}{2} (E - \mu) \right)} \exp \left[ 2i\pi pn(E) \right] \right] E.$$

(11)

where $n(E) = S(E)/b - \gamma$, $S(E)$ being given by the expansion Eq. (6) around the Fermi energy. The next step is to perform the integration around the saddle point $E = \mu$ at low temperature, using the variable $x = \beta(E - \mu)/2\pi$ and replacing $n(E)$ by $n(x)$

$$n(x) = -\gamma + \frac{S(\mu)}{b} + \frac{2\pi x}{\beta \hbar \omega_c} + \frac{2\pi^2 x^2 \tilde{\kappa}}{\beta^2 \hbar \omega_c}.$$  

(12)

In the case of a parabolic band, the first and second terms are related to the Onsager phase or Maslov index, and the oscillation frequency $F = \hbar S(\mu)/e$, respectively. More specifically, magnetization is obtained by differentiation of Eq. (11) with respect to minus $B$, yielding

$$m_{osc} \simeq \frac{e^2 F}{\pi m^*} \sum_{p=1}^{\infty} \frac{A_p}{2\pi p} \sin \left[ 2\pi p \frac{F}{B} - 2\pi \gamma p + \phi_p \right],$$

(13)
where \( A_p \) and \( \phi_p \) are respectively the amplitude and phase of the imaginary damping factor \( Z_p \) defined by

\[
Z_p = A_p \exp(i\phi_p) = \frac{\pi}{2} \int_{-\mu/(2\pi)}^{+\infty} \frac{1 + \tau x/\sigma + \omega x^2/\sigma}{1 + 2\omega x/\tau} \exp\left[2i\pi p x (\tau + \omega x)\right] \cosh^2(\pi x) d\chi, \tag{14}
\]

and which involves the dimensionless parameters

\[
\sigma = \frac{F}{B}, \quad \tau = \frac{2\pi}{\beta \hbar \omega c}, \quad \omega = \frac{\pi \tau \tilde{\kappa}}{\beta}. \tag{15}
\]

Hence, the temperature dependence is given both by \( \tau \propto T \) and \( \omega \propto T^2 \) while \( \omega \), which is proportional to \( S''(E) \), includes the band curvature contribution. The integral can be computed by extending the lower bound to \(-\infty\) at low temperature and by neglecting the factors in front of the exponential that are proportional to parameter \( \sigma^{-1} \) which is generally small compared to unity for magnetic fields strength currently available, except for very small orbits area. Besides, the denominator \((1 + 2\pi x \tilde{\kappa}/\beta)\) is strictly positive as long as \( \tilde{\kappa} \mu < 1 \). In the peculiar case where \( \tilde{\kappa} = 1/\mu \), which is relevant for Dirac fermions, as developed below, we can show that \( \omega = \tau^2/4\sigma \) and that the overall factor reduces actually to \( 1 + \tau x/(2\sigma) \) with no divergence around the lower bound of integration. Following the LK theory and using the residue theorem, we can consider an integration path that goes over the upper complex plane when \( x \) is complex. Indeed, the singularities of the \( \cosh(\pi x)^{-2} \) function give the main contributions to the integral since they are located on the positive imaginary plane \( x_n = i(n + \frac{1}{2}) \). The oscillation amplitude can be written as an infinite summation over the \( x_n \)'s

\[
Z_p = \sum_{n=0}^{\infty} \left\{ 2\pi p \tau + 4i\pi p \omega \left(n + \frac{1}{2}\right) \right\} \exp \left[-2\pi p \tau \left(n + \frac{1}{2}\right) - 2i\pi p \omega \left(n + \frac{1}{2}\right)^2 \right]. \tag{16}
\]

For \( \tilde{\kappa} = 0 \) (or \( \omega = 0 \)), the summation can be performed and we obtain the well known LK thermal reduction factor \( Z_p = R_p = p \pi \tau / \sinh(p \pi \tau) \) \[20\], to which the Dingle factor and, eventually, magnetic breakdown probabilities are added. Eq. \( \tag{16} \) can be expanded up to the first order in \( \omega \propto \tilde{\kappa} \), the sum over \( n \) performed, and the result re-exponentiated, which allows us to rewrite Eq. \( \tag{16} \) as

\[
Z_p \simeq \frac{p \pi \tau}{\sinh(p \pi \tau)} \exp \left[i \frac{\omega}{2\tau} \varphi(p \pi \tau) \right] = R_p \exp \left[i \frac{\pi}{2} \tilde{\kappa} k_B T \varphi \left(2\pi^2 p k_B T / \hbar \omega_c\right) \right], \tag{16}
\]

where

\[
\varphi(x) = \left[ \frac{\sinh(2x) - x - x \cosh^2(x)}{\sinh^2(x)} \right]. \tag{17}
\]
We can identify the amplitude $A_p$ with $R_p$ at this order. As a consequence, while the oscillation amplitude, which is accounted for by the real part of $Z_p$, is unaffected by deviations from parabolicity, an extra phase is present in addition to the $\gamma$ constant in Eq. (13), and which is given by the complex argument. The expression of this additional phase is given by Eq. (1) in the introduction, and is field- and temperature-dependent and proportional to the curvature factor $\kappa$. It is worthwhile to notice that $\varphi(x)$ changes its sign at $x = 1.606115$. Besides, when $x$ is large, i.e. $B/T$ goes to zero, $\varphi(x) \simeq 2 - x$. Nevertheless $x$ is much larger than 2 in this range, hence $\varphi(x) \simeq -x$. Therefore, the phase factor is given by

$$\phi_p \simeq -p \frac{2\pi^4 k_B^2 \hbar}{e} S''(E) \frac{T^2}{B}. \quad (18)$$

In the opposite case, for very large $B/T$ ratio or very small effective mass, $\varphi(x) \simeq x/3$, and the Onsager phase can be approximated by

$$\phi_p \simeq p \frac{2\pi^4 k_B^2 \hbar}{3e} S''(E) \frac{T^2}{B}. \quad (19)$$

The above asymptotic expressions Eq. (18) and Eq. (19) indicate a square temperature dependence and an inverse field dependence of $\phi_p$ both in the high an low field ranges. Besides, within the tight binding model, $S''(\mu) = A_0 m^*^2 / (8\pi \hbar^4)$, indicating largest effect for large unit cell and effective mass. These two features are achieved in organic metals. As an example, unit cell area in the conducting plane as large as $108 \AA^2$ and effective masses in the range $m_\alpha = 3$ to $3.51$ have been reported for the $\alpha$ orbit of $\kappa-(ET)_2[Cu(NCS)]_2$ (where ET stands for the bis-ethelyne-dithio-tetrathia-fulvalene molecule). $\kappa$ can be estimated from band structure calculations and crystallographic data of $\kappa-(ET)_2[Cu(NCS)]_2$, yielding e.g. $\kappa \sim 5 \times 10^{58} \text{m}^{-2} \text{J}^{-2}$ in the $M - \Gamma$ direction, relevant to the $\alpha$ orbit.

As displayed in Fig. 1, we have plotted the Onsager phase $\phi_1$ versus field for several temperature values. The phase is very small at high field. Oppositely, large effects can be observed at low field and high temperature. The dashed line determines the threshold limit when the Landau level gap is equal to the thermal fluctuations, $\hbar \omega_c \simeq k_B T$, and above which the oscillations can be observed at higher fields. Additionally, there is another limit based on the Dingle temperature $T_D$, which also imposes a minimum threshold for the field above which the oscillations can be seen. If we take $T_D = 0.5K$ (the best temperature for $\kappa$-CuNCs) one obtains a threshold field equal to $7.5$ T when $\omega_c \tau = 1$, and where $\tau = \hbar/k_B T_D$ is the scattering time. One however observes oscillations below this threshold.
FIG. 1. Field dependence of the Onsager phase $\phi_1$ at various temperatures deduced from Eq. (1) for parameters relevant to the $\alpha$ orbit of the quasi-two dimensional organic metals $\kappa$-(ET)$_2$[Cu(NCS)$_2$] ($\partial^2 S/\partial E^2 = 5 \times 10^{58} m^{-2} J^{-2}$ and $m^* = 3.2 m_e$). The dashed line marks the field values such as $k_B T \simeq \hbar \omega_c$, which can be regarded as an estimation of the lower limit for the field above which oscillations can be observed.

To our knowledge, quantum oscillations in organic metals have been up to now observed above several teslas at liquid helium temperatures. Therefore, the studied effect could only be observed provided clean compounds with very small scattering rate are synthesized. In this low field range, where $\phi_1/2\pi$ varies by several units, $\phi_p$ can be rewritten as $\phi_p = -2\pi p \delta F/B$ where $\delta F$ is given by:

$$\delta F = \frac{2\pi^3}{\hbar e} S''(\mu)(k_B T)^2. \quad (20)$$
As a typical example, the tight binding model yields

$$\delta F = (k_B T)^2 (\pi^2/24) m^* A_0 / (e \hbar^3).$$

For $m^* = m_e$ and $A_0 = 1 \text{Å}^2$, i.e. for parameters close to those of elemental metals, $\delta F = 3.462 \times 10^{-6} T^2$ which remains negligibly small, even at high temperature. For the above considered organic metal, temperature as high as few tens of a Kelvin is nonetheless necessary to get a frequency shift of few T. Namely, $\delta F$ value as low as 1.5 T should be observed at 20 K which could hardly be detected owing to the oscillation frequency value $F_\alpha = 600$ T yielding $\delta F/F = 0.25\%$.

### III. DIRAC FERMIONS

In this section, we consider the case of Dirac fermions such as observed in monolayer graphene which has been intensively studied (for a review, see e.g. Refs. [30, 31]). The data are in agreement with a linear band dispersion, the curvature parameter being given by $S''(E) = 2 \pi / (\hbar v_F)^2$ with a Fermi velocity $v_F = 10^6 \text{ms}^{-1}$ [18, 32, 33]. Shubnikov-de Haas (SdH) oscillations with effective mass in the range $7 \times 10^{-3} m_e$ to 0.04 $m_e$, depending on the carrier concentration driven by bias voltage, have been observed at temperatures either up to 50 K [18] or above 100 K [32], allowing to expect detection of larger values of the phase $\phi_1$.

Contrary to the parabolic case, the Landau level energies $E_n$ are not linear with index $n$ but are given by $E_n = E_1 \sqrt{n}$, with $E_1^2 = 2e\hbar v_F^2 B$. Since $E_n' \propto n^{-1/2}$, $E_n'' \propto n^{-1}$ and $E_n''' \propto n^{-3/2}$, the two terms in the right hand side of Eq. (5) are diverging when $n$ goes to zero. Therefore, the intermediate step to compute $\Omega_{osc}$ using two integrations by parts in Eq. (5) cannot be performed here, since these contributions makes the integral in Eq. (5) divergent. Taking into account the band degeneracy $g_D = 4$ in the case of graphene, we can write instead

$$\frac{\Omega_{osc}}{A} = -2 g_D b \text{Re} \sum_{p=1}^{\infty} \int_0^\infty \frac{\exp(2i\pi pn)}{1 + \exp[\beta (E_n - \mu)]} E_n' n \frac{E_n''}{2i\pi p} \frac{\exp(2i\pi p E_n^2/E_1^2)}{1 + \exp[\beta (E - \mu)]} E.$$

(21)
An integration by parts is possible if we integrate \( \exp(2i\pi p E^2/E_1^2) \) using the error function in the complex plane

\[
\frac{\Omega_{\text{osc}}}{A} = 2g_D b \Re \sum_{p=1}^{\infty} \frac{\beta E_1}{4(-2i\pi p)^{3/2}} \sqrt{\frac{\pi}{2}} \int_{0}^{\infty} \frac{\text{erf} \left( \sqrt{-2i\pi p E/E_1} \right)}{\cosh^2[\beta(E - \mu)/2]} \, E. \tag{22}
\]

We can set as before \( x = \beta(E - \mu)/2\pi \) and obtain

\[
\frac{\Omega_{\text{osc}}}{A} = 2g_D b \Re \sum_{p=1}^{\infty} \frac{E_1}{2(-2i\pi p)^{3/2}} \frac{\pi^{3/2}}{2} \int_{-\beta\mu/(2\pi)}^{\infty} \frac{\text{erf} \left( \sqrt{-2i\pi p \left( \mu + 2\pi x / \beta \right) / E_1} \right)}{\cosh^2(\pi x)} \, x. \tag{23}
\]

The dominant part of the magnetization is obtained by differentiation of the \( \text{erf} \) function with respect to minus \( B \) in the previous expression, since the dominant oscillating term comes from this function

\[
m_{\text{osc}} = -g_D \frac{e\mu}{\hbar} \Re \sum_{p \geq 1} \frac{\exp(2i\pi p F/B)}{2i\pi p} Z_p,
\]

\[
Z_p = \frac{\pi}{2} \int_{-2\sigma/\tau}^{2\sigma/\tau} \left( 1 + \frac{\tau x}{2\sigma} \right) \frac{\exp(2i\pi p[\tau x + \tau^2 x^2/(4\sigma)])}{\cosh^2(\pi x)} \, x. \tag{24}
\]

\( Z_p \) is normalized such that \( Z_p = 1 \) at zero temperature, i.e. \( \tau = 0 \), using the integral value \( \pi \int_{0}^{\infty} \cosh(\pi x)^{-2} \, x = 1 \). In this expression, the dominant frequency \( F \) is defined by \( \mu^2/E_1^2 = F/B = \sigma \), and the damping factor \( \tau = 4\pi\mu/(\beta E_1^2) \) which is temperature- and field-dependent. These parameters are expressed as

\[
F = \frac{\mu^2}{2e\hbar v_F^2}, \quad \tau = \frac{2\pi\mu}{\beta e\hbar v_F^2 B}, \quad \sigma = \frac{F}{B}, \quad \frac{\tau}{\sigma} = \frac{4\pi}{\beta \mu}. \tag{25}
\]

In reference [34], the authors obtained the expression for the zero-temperature thermodynamic potential, given here by Eq. (23), in terms of a series involving Bernoulli polynomials, up to the fourth power in magnetic field. This is very similar to the expansion obtained for the general case from Eqs. (6) and (11) in the sense that curvature induces an expansion over the cyclotronic frequency. As in Eq. (11), Bernoulli polynomials \( B_n(x) \) of order \( n \) are periodic functions of \( x \) with harmonics \( p \) that decay like \( p^{-n} \). In Eq. (11) this corresponds to an expansion up to \( n = 3 \) that includes the temperature dependence. In the case of Dirac fermions, this expansion is hidden in the factor \( Z_p \) whose dependence on the harmonics index \( p \) can be explicitly determined by expanding the \( x^2 \) argument in the exponential of Eq. (24) as a series. The remaining integrals can in principle be performed in the complex plane yielding a power series in terms of \( 1/p \). In the limit where \( \sigma \gg \tau \), we have
\[ Z_p = \pi p \tau / \sinh(\pi p \tau) = R_p(\pi \tau). \] 
We may then identify \(\pi \tau\) with \(2\pi^2/(\beta \hbar \omega_c)\) as in Eq. \((15)\), by defining the cyclotronic frequency \(\omega_c = eB/m^*\) and effective relativistic mass \(m^* = \mu/v_F^2\).

Otherwise, the amplitude factor \(Z_p = A_p \exp(i\phi_p)\) is complex and possesses a modulus \(A_p\) and non-zero argument \(\phi_p\) contributing to the global Onsager phase of the oscillations.

We may apply the residue theorem to the previous integral, in the limit where the lower bound \(-2\sigma/\tau\) is large, or typically \(T < m^*v_F^2/k_B\). The singularities of the function \(\cosh(x)^{-2}\) are still located at \(x_n = i(n + \frac{1}{2})\) and we obtain

\[ Z_p \simeq \sum_{n \geq 0} \left\{ 2p\pi \tau \left[ 1 + \frac{i\tau(n + \frac{1}{2})}{2\sigma} \right]^2 + \frac{\tau}{2i\sigma} \right\} \exp \left[ -2\pi p \tau(n + \frac{1}{2}) - 2i\pi p \frac{\tau^2}{4\sigma}(n + \frac{1}{2})^2 \right]. \tag{26} \]

**FIG. 2.** Field dependence of the Onsager phase \(\phi_1\) for Dirac fermions derived from Eq. \((2)\) with Fermi velocity \(v_F = 10^6\text{ms}^{-1}\) and effective mass \(m^* = 0.02\) at \(T = 10\text{ K}, 50\text{ K} \text{ and } 100\text{ K}\).

This expression can be compared to Eq. \((16)\) provided substitutions \(\tilde{\kappa} = 1/\mu\) and \(\omega = \tau^2/4\sigma\) are made. Except for the argument of the exponential terms, Eq. \((26)\) is different from
the general case, yielding therefore a modified form of the Onsager phase $\phi_p$ and function $\varphi(x)$ defined in Eq. (17), given by Eq. (2). In this case, the series expansion valid at low $m^*T/B$ ratio gives $\varphi(x) \simeq x$ instead of $x/3$, and the phase $\phi_p$ is inversely proportional to the magnetic field. The other limit as $m^*T/B$ is large, is as previously negative, although $\varphi(x) \simeq 4 - x$ instead of $2 - x$. As in the previous case, $x$ is much larger than 4 in this range, hence $\varphi(x) \simeq -x$. Therefore, the phase factor is given by

$$
\phi_p = \pm \frac{2\pi^3 p}{\hbar e B} \left( \frac{k_B T}{v_F} \right)^2,
$$

(27)

where the sign plus and minus corresponds to the high and low field limit, respectively. According to Eq. (27), $\phi_p$ depends on the Fermi velocity, instead of the effective mass and band curvature, at both high and low field. Field dependence of $\phi_1$ is displayed in Fig. 2 for experimental parameters relevant to graphene [18, 32]. As reported in the preceding section, negligibly small values are obtained in the high field range. In the low field range, the same behaviour as for organic metals is obtained. Namely, a significant drop down of $\phi_1$ at low field and high temperature. Nevertheless, the frequency shift, which can be written $\delta F/F = 2\pi^2 \left( \frac{k_B T}{m^* v_F} \right)^2$, remains small (about 8 % at 100 K in the example of Fig. 2) although higher than in the case of the organic metal considered in the preceding section.

IV. SUMMARY AND CONCLUSION

We have evidenced that a non-parabolicity of the band dispersion versus momentum yield a field-dependent shift of the Onsager phase of quantum oscillations, i.e. a frequency shift. The analytical expressions of this phase shift depend on the nature of the dispersion band, either mainly parabolic with corrective terms to the parabolicity (organic conductors) or linear (Dirac fermions). It is demonstrated that, in both cases, the effect is amplified at low magnetic field, high temperature and either large effective mass and unit cell area (organic conductors) or small Fermi velocity (Dirac fermions). However, the observed effect is small within the experimental conditions where quantum oscillations are observed. Largest effect are nevertheless observed for Dirac fermions. Indeed, the deviation from parabolicity of their band dispersion is the most significant. Since the phase shift is expected to be the largest for small Fermi velocity, Dirac fermions in organic conductors, such as observed in $\alpha$-(BEDT-TTF)$_2$I$_3$ [35, 36], with large unit cell, hence small Fermi velocity appear as
promising candidates. It is expected that samples should be clean enough to observe the large phase shift deviations at low fields. This typically corresponds to the integer quantum Hall regime, which requires a proper treatment of the disorder effects but are not that relevant for the magnetization oscillations, in contrast to transport coefficients. The temperature-dependent phase shifts are not expected to be fundamentally affected. Indeed, the principal modification for the magnetization oscillations in the quantum Hall regime will take place at the level of the disorder Dingle factor (associated with a Lorentzian distribution of the Landau bands) and may be no more valid in low-dimensions, and can be typically replaced by a Gaussian reduction factor (with a Gaussian dependence on the harmonics $p$), which reflects a Gaussian shape of the Landau bands as often reported experimentally for the density of states in the integral quantum Hall regime [37]. Also, it may be important to consider theoretical Fermi surfaces with singular behavior which is affected by large curvature and therefore may display important phase shifts.

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