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Suppression of field-induced chemical potential oscillations in ideal two-bands compensated metals

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The field and temperature dependence of the de Haas-van Alphen oscillations spectrum is studied for an ideal two-dimensional compensated metal. It is shown that the chemical potential oscillation, otherwise involved in the frequency combinations observed in the case of uncompensated orbits, is strongly damped and even suppressed in the case where the effective mass of the electron- and hole-type orbits are the same. This damping is even more pronounced in the case where magnetic breakdown occurs.

INTRODUCTION

In high enough magnetic fields, the Fermi surface (FS) of multiband quasi-two-dimensional metals, are liable to give rise to networks of orbits coupled by magnetic breakdown (MB). The most studied type of network is the linear chain of coupled orbits introduced by Pippard [1] and illustrated by several quasi-two-dimensional (q-2D) organic conductors such as κ -(BEDT-TTF)₂Cu(NCS)₂. As discussed, in Ref. [2], magnetic oscillations spectra of such networks contain many frequencies that are linear combinations of two basic frequencies. In addition to those linked to MB-induced orbits, other frequencies are observed that are not accounted for by the semi-classical theory of Falicov and Stachowiak [3]. They can be attributed to quantum interference (as far as magnetoresistance oscillations are concerned), MB-induced modulation of the density of states [4, 5, 6] and oscillation of the chemical potential [7, 8, 9, 10], even though the actual respective contribution of these three phenomena to the oscillatory behavior remain to be established.

Another type of network is provided by quasi-two-dimensional (q-2D) metals of which the FS is composed of compensated electron- and hole-types tubes. This is the case of organic metals of which the FS originates from the overlapping in two directions of hole tubes with an area equal to that of the First Brillouin zone (FBZ) and from the resulting gap openings [11]. Such networks are also observed in the family of organic metals (BEDT-TTF)₈Hg₄Cl₁₂(C₆H₅X)₂ (X = Cl, Br) whose FS, which originates from two pairs of crossing q-1D sheets, is composed of one electron and one hole tube with the same area [12]. As it is the case of the above-mentioned linear chains of coupled orbits, magnetoresistance oscillations spectra in this type of network reveal frequencies that are linear combinations of three basic frequencies, linked to the compensated orbits and to the two FS pieces located in-between [13, 14, 15]. However, in striking

contrast to the data relevant to linear chains of orbits, de Haas-van Alphen (dHvA) oscillations spectra recorded in the case of the compound with X = Br only exhibit oscillations, the field and temperature dependence of which can be consistently interpreted on the basis of the semiclassical model of Falicov and Stachowiak [3, 15].

The aim of this paper is to explore the field and temperature dependence of the dHvA oscillations spectra of an ideal 2D metal whose FS is composed of one electron- and one hole-type compensated orbit. It is demonstrated that the field-induced oscillation of the chemical potential is strongly damped for such a FS and even suppressed in the case where electron- and hole-type orbits have the same effective mass. In addition, the chemical potential oscillation is even more damped in the case where the two orbits are coupled by MB.

MODEL

We consider a 2D metal whose electronic structure consist of two parabolic bands of hole- and electron-type, respectively. The bottom of the electron band is set at zero energy while the top of the hole band is at $\Delta > 0$. The total number of electrons in the system is such that the hole band is completely filled. Since the lower part of the electron band is lower in energy than the top of the hole band, some quasiparticles move to the electron band in order to lower the total energy. The effective masses linked to the two bands, $m_e^* \equiv 1/C_e$ and $m_h^* \equiv 1/C_h > 0$, can be different. It is useful to define the physical units of the problem. The reduced field $b = eBA_0/h$ is in units of the characteristic field $\tilde{B} = h/eA_0$, the effective masses are in units of the electron mass m_e , the energies are in units of $\tilde{E} = 2\pi\hbar^2/m_eA_0$ and the temperature t in units of $\tilde{T} = \tilde{E}/k_B$. Given a unit cell area $A_0 = 197.6 \text{ \AA}^2$, which stands for (BEDT-TTF)₈Hg₄Cl₁₂(C₆H₅Br)₂ [16], we obtain $\tilde{B} = 2093 \text{ T}$ and $\tilde{T} = 2812 \text{ K}$. Therefore, realistic experimental conditions yield small values

of b and t compared to \tilde{B} and \tilde{T} , respectively. The semiclassical quantization of the energy levels in the presence of a magnetic field leads to the Landau equations:

$$E_e(n) = C_e b(n + \frac{1}{2}), \quad E_h(n) = \Delta - C_h b(n + \frac{1}{2}) \quad (1)$$

each Landau level (LL) having a degeneracy b per sample area. The zero field Fermi energy is simply given by $E_F = m_h^*/(m_e^* + m_h^*)\Delta$. At finite temperature, the total free energy is given by the difference between the contribution of the electron and of the hole band, with the condition that the total number of quasiparticles N_t is fixed. In addition, the compensation condition imposes that the number of quasiparticles in the electron (N_e) and in the hole (N_h) band are the same. From the thermodynamical relations, we can define a free energy for the system based on the difference between the free energy for the electrons in the electron band and the free energy for the holes in the hole band $\Delta F = \Omega_e - \Omega_h + (N_e - N_h)\mu$, where $\Omega_{e(h)}$ is the Grand Potential for the electrons (holes) and μ is the chemical potential [$\mu(T=0) = E_F(B)$]:

$$\Omega_e(T, B) = -Tb \sum_{n \geq 0} \log(1 + \exp[\beta(\mu - E_e(n))])$$

$$\Omega_h(T, B) = Tb \sum_{n \geq 0} \log(1 + \exp[\beta(E_h(n) - \mu)]) \quad (2)$$

Since $N_e = N_h$, we can conclude that $F = \Omega_e - \Omega_h$ in compensated metals. μ is evaluated from the self-consistent equation $\partial \Delta F / \partial \mu = 0$. At zero temperature, the above expressions reduce simply to the ground state (GS) energy ΔE_0 . The Fermi energy $E_F(B)$ is given by the condition $n_e b = n_h b$, where n_e and n_h are the number of LL filled, which leads to $n_e = n_h$, and

$$\begin{aligned} \Delta E_0 &= -b \sum_{n=0}^{n_e-1} (E_F - E_e(n)) - b \sum_{n=0}^{n_h-1} (E_h(n) - E_F) \\ &= E_e - E_h + b(n_h - n_e)E_F = E_e - E_h \end{aligned} \quad (3)$$

The special cases where the Fermi energy goes through one Landau level, and where this Landau level is partially filled, correspond to singular points in the energy spectrum as a function of the inverse field that do not modify the thermodynamical quantities. The exact expression for ΔE_0 is simply

$$\Delta E_0 = (C_e + C_h)b^2 n_e^2 - b \Delta n_e \quad (4)$$

with $n_e = [E_F/C_e b + 1/2]$. We deduce the oscillating part of the magnetization $M_{osc} = -\partial \Delta E_0 / \partial b$ from the latter expression:

$$M_{osc} = -(C_e + C_h)F_0^2 \sum_{k \geq 1} \frac{(-1)^k}{\pi k} \sin(2\pi k F_0/b) \quad (5)$$

where $F_0 = m_e^* m_h^* \Delta / (m_e^* + m_h^*)$ is the fundamental frequency corresponding to the FS area of the electron and hole band. At zero temperature and for fixed number of electrons, the magnetization oscillates with the frequency F_0 , and the amplitude given by the Lifshits-Kosevich (LK) formula with a $1/k$ dependence of the k^{th} harmonics [17].

SELF-CONSISTENT EQUATION FOR THE CHEMICAL POTENTIAL

The oscillatory part of the Grand Potentials can be extracted from Eqs. (2) using Poisson's formula (see Ref. [17]):

$$\Omega_e \simeq -\frac{1}{2C_e} \mu^2 + \frac{b^2 C_e}{2} \left[\frac{1}{12} + \sum_{n=1}^{\infty} \frac{(-1)^n}{\pi^2 n^2} R_{e,n} \cos(2\pi n \frac{\mu}{C_e b}) \right]$$

$$\Omega_h \simeq \frac{1}{2C_h} (\Delta - \mu)^2 \quad (6)$$

$$- \frac{b^2 C_h}{2} \left[\frac{1}{12} + \sum_{n=1}^{\infty} \frac{(-1)^n}{\pi^2 n^2} R_{h,n} \cos(2\pi n \frac{\Delta - \mu}{C_h b}) \right] \quad (7)$$

where $R_{e(h),n} = 2\pi^2 n m_{e(h)}^* t / [b \sinh(2\pi^2 n m_{e(h)}^* t/b)]$ are the reduction factors. The chemical potential satisfies therefore the self-consistent relation:

$$\begin{aligned} \mu &= E_F + \frac{b}{m_e^* + m_h^*} \sum_{n=1}^{\infty} \frac{(-1)^n}{\pi n} [R_{h,n} \cos(2\pi n \frac{\Delta - \mu}{C_h b}) \\ &\quad - R_{e,n} \cos(2\pi n \frac{\mu}{C_e b})] \end{aligned} \quad (8)$$

At a first order approximation, we can replace μ in the previous expression by E_F since the second term is proportional to b , which is small compared to E_F . This gives:

$$\mu \simeq E_F + \frac{b}{m_e^* + m_h^*} \sum_{n=1}^{\infty} \frac{(-1)^n}{\pi n} (R_{h,n} - R_{e,n}) \cos(2\pi n \frac{F_0}{b}) \quad (9)$$

It can be remarked first that, in the case where the effective masses linked to the electron- and hole-type orbits are the same, the oscillatory part of Eq. 9 vanishes and the chemical potential remains constant in magnetic field. More generally, the amplitude of the chemical potential oscillation can be compared to the case of two electron orbits [10]. In Fig. 1, the field-dependent chemical potential is calculated for compensated orbits with

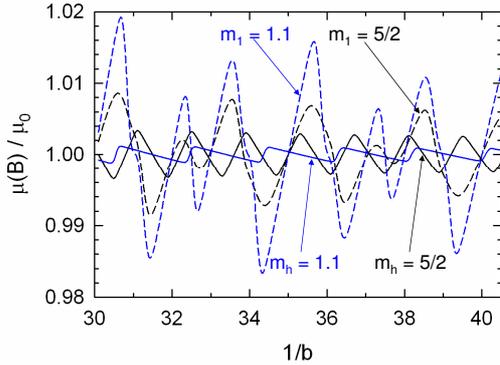


FIG. 1: (color online) Field dependence of the chemical potential normalized to its value in zero-field at $t = 0.001$. Solid and dashed lines correspond to one electron and one hole compensated orbits (with $m_e^* = 1$ and $m_h^* = 1.1$ or $5/2$) and two electron orbits (with $m_0^* = 1$ and $m_1^* = 1.1$ or $5/2$), respectively (see text).

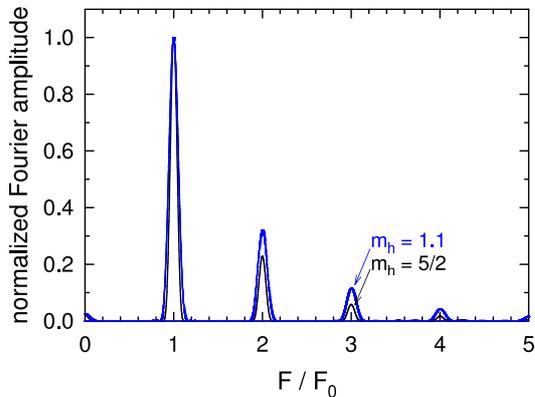


FIG. 2: (color online) Fourier spectrum of the magnetization at $t = 0.001$ for $m_e^* = 1$ and $m_h^* = 1.1$ or $5/2$. The field range is from $b = 0.015$ to $b = 0.033$. F_0 is the fundamental frequency (see text).

$m_e^* = 1$ and $m_h^* = 1.1$ or $5/2$ and compared to the case of two electronic orbits with effective masses $m_0^* = 1$ and $m_1^* = 1.1$ or $5/2$. It can be observed that the chemical potential oscillations are strongly damped for compensated orbits, even in the case where m_h^* and m_e^* have strongly different values [18].

DE HAAS-VAN ALPHEN OSCILLATIONS

The oscillatory magnetization can be obtained putting the expression of the chemical potential given by Eq. (9) back into the expression for the free energy F using Eqs. (2). As above discussed, the chemical potential is field-independent for $m_e = m_h$ and the Lifshits-Kosevich (LK) formula [17] holds in this case. However, for $m_e^* \neq m_h^*$, the oscillatory magnetization differs from

the LK theory. Examples of Fourier analysis of the magnetization are given in Fig. 2. In addition to the absence of frequency combinations in the Fourier spectra, the striking feature is the large amount of harmonics.

It is useful for experimentalists, to check to what extent it is possible to determine a temperature and field range in which the LK formalism provides a satisfactory approximation of the oscillatory behavior in the case where $m_e^* \neq m_h^*$. The b/t dependence of the magnetization with frequencies F_0 and $2F_0$ are given in Fig. 3. The LK formula satisfactorily account for the field and temperature dependence of the first harmonics. Furthermore, it can be noticed that in the case where m_h^* is strongly different from m_e^* , the contribution of the orbit with the smallest effective mass dominates in a large field range (see the dashed line in Fig. 3a). It has also been checked that, in the opposite case where m_e^* is close to m_h^* , the data can be accounted for by the contribution of only one orbit with a mean effective mass, namely, $m_{mean}^* \simeq (m_e^* + m_h^*)/2$. In contrast, the LK formula cannot account for the second harmonics in the case where m_e^* and m_h^* significantly differ. The observed behavior of the second harmonics in this latter case is reminiscent to that observed for two electronic orbits [10].

MAGNETIC BREAKDOWN

Provided the magnetic field is high enough to overcome the MB gap, combinations of different orbits can be observed. Assuming that p and q are the probability amplitude of tunnelling and of staying on the same band yields $p^2 + q^2 = 1$ [3, 17]. Depending on the band type (electron or hole), a quasiparticle acquires a phase either $\sigma_e = S_e/2\pi b$ or $\sigma_h = S_h/2\pi b$. The semiclassical actions are given by $S_e = 2\pi m_e^* E > 0$ and $S_h = 2\pi m_h^* (\Delta - E) > 0$. Taking into account the turning points and the chirality of the orbits, the LLs are now solution of the implicit equation:

$$(1 + q \exp i\sigma_e)(1 + q \exp -i\sigma_h) + p^2 \exp i(\sigma_e - \sigma_h) = 0 \quad (10)$$

In the case where the bands are independent ($p = 0$), Eq. (10) can be factorized and reduces simply to $1 + \exp(i\sigma_e) = 0$ or $1 + \exp(-i\sigma_h) = 0$, which corresponds to the two independent sets of levels given by Eq. (1). Eq. (10) can be solved numerically for $p \neq 0$: the field dependence of the LLs energy is given in Fig. 4 in the high field range for $q = 0.8$. For $p \neq 0$, gaps open at the former LL intersections, and the structure of the two bands is modified. Alike the case where $p = 0$, a quasi-hole and a quasi-electron band can be defined. Indeed, levels that

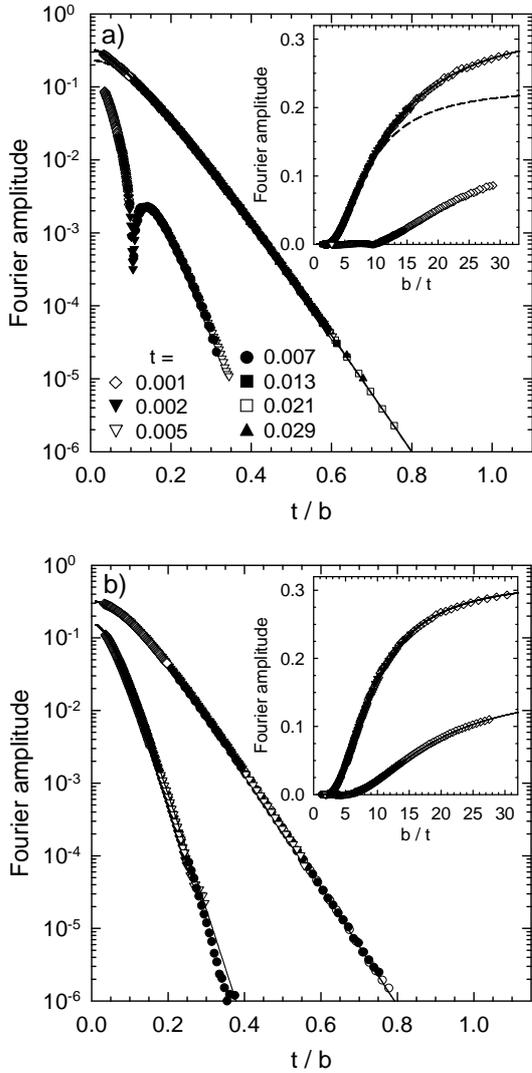


FIG. 3: t/b dependence of the Fourier components of the magnetization with frequencies F_0 and $2F_0$. The effective mass of the electronic-type orbit is $m_e^* = 1$. The effective mass of the hole-type orbit is $m_h^* = 5/2$ and 1.1 in (a) and (b), respectively. Solid lines are obtained from the LK formula. The dashed line in (a) corresponds to the contribution of the electronic orbit.

go upwards above $E = E_F(B)$ (see red solid lines in Fig. 4) and downwards below $E_F(B)$ (see black solid lines) with the field variation can represent quasi-electron and quasi-hole bands, respectively. At zero temperature, the compensation condition $N_e = N_h$ is replaced by the condition that the lower quasi-hole band is filled completely, the upper electron-band being empty. The two bands are therefore separated by the chemical potential, as shown in Fig. 4 where the Fermi energy in zero-field (E_F) as well as the field-dependent Fermi energy ($E_F(B)$) are also plotted. As the magnetic field decreases, the field-dependent Landau levels near the Fermi energy are flatter since the levels of the two bands can not cross the line

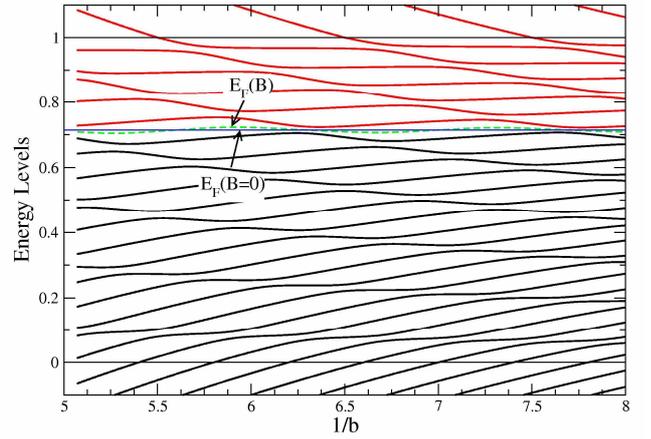


FIG. 4: (color online) Energy levels for a reflection amplitude $q = 0.8$ (or, equivalently, tunnelling amplitude $p = \sqrt{1 - q^2}$) $\Delta = 1$, $m_e^* = 1$ and $m_h^* = 5/2$. Levels that belong to a quasi-electron band (going upwards when the field increases, red lines) above the Fermi energy can be distinguished from levels that belong to a quasi-hole band (respectively going downwards, black lines). Above Δ the LLs correspond to the electron band only, with energies $E_e(n)$, whereas LL $E_h(n)$ describe the negative sector.

defined by the chemical potential.

In order to estimate the total free energy, it is necessary to modelize the lower quasi-hole band since the GS energy ΔE_0 given by Eq. (3) is no more well defined, due to the mixing between electron and hole levels. In particular, the GS energy should correspond to the sum of all the quasi-hole energies below the Fermi energy $E_F(B)$. However, in our model, the energies are not bounded by below and the GS energy is formally infinite. We propose to account for this problem by fixing the number N_L of quasi-hole LLs and incorporate formally a reservoir of electrons, in such a way that the total number of particles is fixed. The reservoir consists in a continuous band between the energies $-E_+ > -E_-$ with $E_{\pm} > 0$. This band is assumed to be situated below the lowest of the N_L^{th} level and its quasi-particle density is fixed to unity. The total and constant quasi-particle number ρ_0 is then given by

$$\rho_0 = E_+ - E_- + N_L b. \quad (11)$$

This gives a relation between E_{\pm} and the field. We can for example either fix E_+ and express E_- as function of b , or conversely. The two cases $q = 1$ and $q = 0$ can be solved exactly: for $q = 1$, the total energy of the hole band $E_h(n)$, when it is completely filled, is

$$E_0 = \Delta b N_L - \frac{1}{2} C_h b^2 N_L^2 + \frac{1}{2} (E_-^2 - E_+^2), \quad (12)$$

and the GS energy E_{GS} when we include the electron band is

$$E_{GS}(q=1) = E_0 + \Delta E_0(q=1) < E_0 \quad (13)$$

where $\Delta E_0(q=1)$ is given by Eq. (4). E_0 is a quadratic function of the field and therefore does not contribute to the magnetic oscillations. In the opposite case when $q=0$, the LL energies between 0 and Δ are easily obtained by solving Eq. (10):

$$E_{eh}(n) = E_F - b \frac{C_e C_h}{C_e + C_h} \left(n + \frac{1}{2} \right) = \frac{C_e}{C_e + C_h} E_h(n) \quad (14)$$

where E_F is the Fermi energy in zero field. Within the semi-classical approach, this means that the cross-section area of the FS corresponding to the parabolic quasi-hole band, which is completely filled, is zero. Since it is proportional to the frequency of the oscillations, these oscillations should vanish as $q=0$. This simple feature is in line with the prediction of the semiclassical model for two compensated orbits in the case where $q=0$, keeping in mind that these orbits have opposite signs [3]. Below energy zero, the LLs given by Eq. (14) join continuously the LL $E_h(n)$ above zero. In our model, there are $n_p = [x\Delta/C_h + \frac{1}{2}]$ levels with positive energies $E_{eh}(n = 0..n_p - 1)$ ($E_{eh}(n_p - 1) > 0 > E_{eh}(n_p)$), and $N_L - n_p$ levels with negative energies $E_h(n = n_p..N_L - 1)$. The total GS energy is then given by the formula

$$\begin{aligned} E_{GS}(q=0) &= E_F b n_p - \frac{1}{2} \frac{C_e C_h}{C_e + C_h} b^2 n_p^2 \\ &+ \Delta b (N_L - n_p) - \frac{1}{2} C_h b^2 (N_L^2 - n_p^2) \\ &+ \frac{1}{2} (E_-^2 - E_+^2). \end{aligned} \quad (15)$$

The last expression can be rewritten as $E_{GS}(q=0) = E_0 + \Delta E_0(q=0)$, with

$$\Delta E_0(q=0) = \frac{C_h}{C_e + C_h} \left(-\Delta b n_p + \frac{1}{2} C_h b^2 n_p^2 \right). \quad (16)$$

In particular, as the magnetic field goes to zero, we obtain $\Delta E_0(q=0) = \Delta E_0(q=1) = -\Delta^2/2(C_e + C_h)$. For $\Delta = 1$, $C_e = 1$ and $C_h = 2/5$, $\Delta E_0 \simeq -0.357$, which is the average value around which the free energy is oscillating. In the general case, $\Delta E_0(q=0)$ oscillates with a frequency equal to $\Delta/C_h > F_0$ which comes from the oscillation of n_p with the field. This frequency gradually appears in the spectrum of the magnetization as q decreases, while the amplitude of the main physical frequency F_0 vanishes with q , as expected. This artefact is due to the way we modelize the bottom of the lower band.

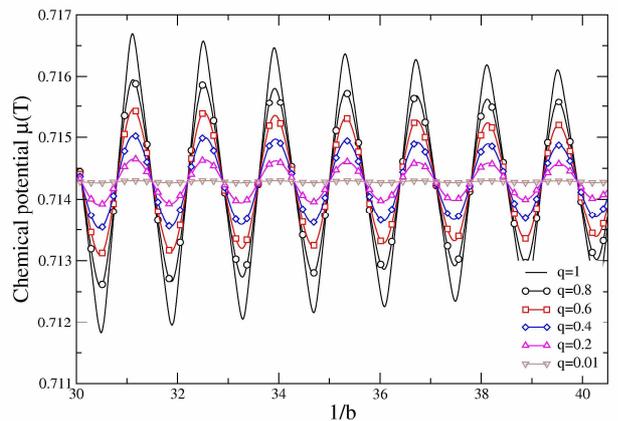


FIG. 5: (color online) Oscillation of the chemical potential at $t = 0.001$ for different values of q ; $\Delta = 1$, $m_e^* = 1$ and $m_h^* = 5/2$.

Indeed at the junction where the LLs cross the zero energy, there is a discontinuity of their slope as function of the field (in Fig. 4 this is more obvious for small values of q) which can be viewed as a discontinuity of the mass since there is no band mixing below the zero energy.

However, the chemical potential is not affected by this artificial oscillation pattern, as we can see in Fig. 5, where the field-dependent variation of the chemical potential has been plotted for different values of q . As q decreases, i.e., as the probability amplitude of tunnelling increases, the chemical potential oscillations decreases until it finally vanishes at $q=0$.

The problem of modeling the lower band junction is probably intrinsic to choosing a parabolic form that correctly describes the physics locally around the FS, but no around the band singularities or special symmetry points in the Fourier space. Indeed, the geometry of the band can change. One example is related to the tight-binding model, or Harper model, with a single band (see [19, 20]). Within this model, an electron can hop from one site to the other with an amplitude t . Without a magnetic field, the energy spectrum is simply $E(k) = -2t[\cos(k_x) + \cos(k_y)]$, which is symmetric around the zero energy by the translation $k_x \rightarrow k_x \pm \pi$ and $k_y \rightarrow k_y \pm \pi$. For small k or low energy, the band is parabolic $E(k) \simeq t(-2 + k^2)$, of electron nature. In the presence of a small magnetic field, a spectrum of LLs with constant gaps is obtained. As the energy increases, the nature of the band changes, and at half filling, $E = 0$, the semiclassical trajectories allowed by the LK theory are open paths $k_y = \pm k_x \pm \pi$, and, a priori, no quantification is possible even if it has been shown that an oscillation of frequency one half of the Brillouin zone area occurs [19], with an amplitude which decreases as the inverse of the logarithm of the field. This frequency corresponds to half of the area of the Fermi surface because the band is half

filled, and can be viewed also as the surface enclosed by the intersections of the straight trajectories. The surface area of the orbits in the Fourier space in this case is

$$S(E) = \frac{4}{\pi^2} \int_{-2}^{E/t} \frac{1}{2-x} K \left[\left(\frac{2+x}{2-x} \right)^2 \right] dx \quad (17)$$

where K is the complete elliptic integral of the first kind $K(m) = \int_0^1 dx / \sqrt{(1-x^2)(1-mx^2)}$. At half filling, the derivative of $S(E)$ diverges, which means that the gap between the LLs vanishes and the mass of the quasi-particle becomes infinite. The singularity at $E = 0$ separates 2 regions of different nature (electron band near $E = -2t$, and hole band near $E = 2t$), but related by a translation symmetry. This example shows that a simple model of parabolic band can not account for singularities occurring in more complicated cases, which can be important to compute the total energy like in E_{GS} , even though it describes correctly quantities, such as the chemical potential, which are not sensitive to singularities far for the FS. A modified version of this tight-binding Hamiltonian including a full 2 band mixing could in principle be used to modelize the singularity at the bottom of the previous quasi-hole band, in order to avoid the complications due to the discontinuities at the junctions when the LLs cross the zero energy.

SUMMARY AND CONCLUSION

The field-dependent chemical potential oscillation in a network of compensated electron and hole orbits is strongly damped when compared to the case of a FS with only electronic orbits [7, 8, 9, 10]. It is even suppressed in the case where the effective mass of electron (m_e) and hole (m_h) band are the same. In addition, the LK formula accounts for the field and temperature dependence of the first harmonic's dHvA oscillations amplitude in all cases. As for the amplitude of the second harmonic, it is accounted for by the LK formula, provided m_e and m_h are not strongly different. Besides, as MB develops, the chemical potential oscillation is further damped. This set of results provide a strong basis for the interpretation of the dHvA spectra of networks of compensated electron and hole orbits that are illustrated by the FS of numerous organic metals. Calculations involving the topology of the FS, which would allow to account for the contribution of the MB-induced modulation of the density of states [4, 5, 6], are now necessary in order to get a com-

prehensive understanding of the magnetic oscillations of these systems.

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