Magnetic oscillations and frequency mixing in a two-band conductor

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Abstract

Exact analytical results of the de Haas-van Alphen (dHvA) effect in an idealized two-band Fermi liquid with parabolic dispersion are presented. We consider a Fermi surface consisting in two electron bands with different band edges and band masses. Magnetic breakthrough (MB) between the bands is negligible. Analytical expressions of the dHvA Fourier amplitudes are derived in the case where the total number of electron is fixed (Canonical Ensemble, CE). As already reported in the literature, the oscillations of the chemical potential yield frequency mixing and Lifshitz-Kosevich (LK) theory, which is valid in the Grand Canonical Ensemble (GCE), does not apply at very low temperature. We show that the corresponding Fourier amplitudes depend on the commensurability between the two effective masses and also the two fundamental frequencies.

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Experimental data on dHvA quantum oscillations in organic conductors show evidence of a deviation with LK [1] and Falikov-Stachowiak (FS) [2] theories for low dimensional systems. We observe in the Fourier spectrum of the magnetization in the transfer salt $\kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2$ [3] frequencies that are not related to a classical orbit on the Fermi surface. For these “forbidden” frequencies, the electron orbits locally in the opposite direction imposed by the magnetic field. This material is made of 2 disconnected Fermi surfaces, one pocket hole and an open one dimensional surface. From LK and FS theories, we should obtain a small frequency $f_\alpha$ corresponding to the pocket, and, by MB effect between the pocket and the open surface, a larger frequency $f_\beta > f_\alpha$ at higher field (>20T at 1K), plus their harmonics and classical combinations that are permitted like $f_\alpha + f_\beta$, $2f_\beta - f_\alpha$ etc... due to MB and Bragg reflections throughout the Brillouin zone. The “forbidden” frequencies seen experimentally correspond to $f_\beta - f_\alpha$, $2f_\beta - 2f_\alpha$. In Shubnikov-de Haas (SdH) transport experiments, they may be

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explained by quantum interference or Stark effect, like in \( (BEDO - TTF)_5CsHg(SCN)_4 \) \[7\]. In dHvA experiments, the explanation seems rather different because Stark effect does not apply. There is an argument \[4,5,6\] showing that fluctuations of the chemical potential are important in these low dimensional systems, especially at very low temperature (<1K) in the high field range. This implies that LK and FS frequencies and Fourier amplitudes should be different in the CE, where the electron density \( n_e \) is fixed, and that the classical way of defining a frequency as area of a classical orbit is incorrect in this ensemble. However, since the LK and FS are correct in the GCE, thermodynamical relations connect the 2 Ensembles. Determinating the chemical potential is not simple in the finite temperature case and a complete calculation is necessary. However we can obtain analytical results at \( T=0 \) in some cases \[6\].

In order to reproduce the main features of transfer salt Fermi surface, we considder a two-band model with gaps \( \Delta_{\alpha=0.1} \) and effective masses \( m^*_\alpha=0.1 \) in an external magnetic field \( B \) (we neglect magnetic interaction). This model has been considered by other authors mostly in a numerical way \[4,8\]. However there is no MB unlike BEDT systems. The Landau levels (LL) of each band \( \alpha = 0, 1 \) have energies:

\[
e_\alpha(n) = \Delta_\alpha + (h^2/2\pi m^*_\alpha)(eB/h)(n + 1/2).
\]

We will define energies in unit of \( 2\pi h^2/m^*_\alpha \) and \( b = eB/h \) is the dimensionless field per unit cell area. In absence of field, the electron densities \( n_\alpha \) of each band satisfy the conservation equation \( \sum \alpha n_\alpha = n_e \). If \( S_\alpha \) is the area of the \( \alpha \)-band Fermi surface, then \( n_\alpha = S_\alpha/4\pi^2 \). In term of the Fermi energy \( E_f \), we have \( S_\alpha = 2\pi m^*_\alpha h^2/(E_f - \Delta_\alpha) \). In the following, we will use the dimensionless variable \( x = n_e/b \), and the ratios \( e_\alpha = m^*_0/m^*_\alpha \). In the new units, the LL are \( e_\alpha(n) = \Delta_\alpha + e_\alpha b(n + 1/2) \). It is also convenient to use the dimensionless fundamental frequencies \( f_\alpha = S_\alpha/4\pi^2 n_e \), with \( f_0 + f_1 = 1 \). Defining \( f_b = (\Delta_0 - \Delta_1)/c_1 n_e \), we obtain

\[
f_0 = \frac{c_1}{1 + c_1}(1 - f_b), \quad \text{and} \quad f_1 = \frac{1}{1 + c_1}(1 + c_1 f_b)
\]

From the LK theory, the Grand Potential \( \Omega \) is the sum of each band contribution \( \Omega_{\alpha} \):

\[
\Omega_{\alpha} = -\frac{1}{2c_\alpha}(\mu - \Delta_\alpha)^2 + \frac{b^2c_\alpha}{2}\left[\frac{1}{12} + \sum_{p \geq 1} \frac{(-1)^p}{\pi^2 p^2} R_{\alpha,p}(T) \cos \left(2\pi p \frac{\mu - \Delta_\alpha}{c_\alpha b}\right)\right]
\]

\( R_{\alpha,p}(T) = p\lambda_\alpha / \sinh(p\lambda_\alpha) \) is the temperature reduction factor, and \( \lambda_\alpha = 2\pi^2 T/c_\alpha b \). The oscillating part of the magnetization \( M = -\partial\Omega/\partial B \) contains cosine functions of arguments \( 2\pi p F_\alpha/B = 2\pi p f_\alpha x \) with \( p \) integer, and therefore the Fourier spectrum is made of the individual frequencies \( F_\alpha \) plus their harmonics. There is no mixing. In the CE, we will show that the oscillating part of the magnetization \( M = -\partial\Omega/\partial B \), where \( F = \Omega + n_\mu \) is the free energy, contains the combinations \( k_f + 1 \). The chemical potential at finite field is the implicit solution of the conservation equation \( \partial\Omega/\partial \mu = -n_e \). Replacing the solution \( \mu \) into Eq. (2) for \( \Omega \), we can obtain the free energy, hence \( M \).

At zero temperature, we can solve the chemical potential equation exactly and obtain the exact total energy \( E \) as a function of \( x \). \( E \) can be expressed as \( E_0 + b^2 G(x)/2 \), with \( G(x) \) an oscillatory function, and \( E_0 \) the zero field energy:

\[
E_0 = \frac{c_1}{2(1 + c_1)} n_e^2 + \frac{n_e}{1 + c_1} (c_1 \Delta_0 + \Delta_1)
- \frac{(\Delta_0 - \Delta_1)^2}{2(1 + c_1)}
\]

We used a combinatorial analysis to compute \( G \). We had to find the \( |x| + 1 \) lowest LL between the 2 bands, with the first \( |x| \) LL completely filled and the last partially. After tedious computations, we arrive at the following expression:

\[
G(x) = \frac{1}{12} \sum_{l=1}^{\infty} \frac{1}{l^2} \cos(2\pi l f_0 + f_1) x
+ \sum_{l \geq 1} \frac{1}{2\pi^4 l^4} \left(1 - c_1 f_0 \right) \cos(2\pi l f_0 x)
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\]
+ 2 \sum_{l \geq 1} \sum_{l' \neq l} \frac{(-1)^{l'-l}}{4\pi^2(l'-l)^2} \frac{(1 + c_1)^2}{(l + l'c_1)^2} \\
\times \left(1 - \cos \frac{2\pi(l' - l)c_1}{1 + c_1} \right) \cos \left(2\pi(l'f_0 + l f_1)x \right).

Eq. (4) is the main result for the two-band problem at zero temperature. The oscillating part of the magnetization is simply \( m_{osc} = n_c G'(x)/2 = \sum F(A(F) \sin(2\pi F x)) \). In the double sum, it can happen that some terms diverge when \( l + l'c_1 \) vanishes in the denominator. But in this case, the quantity \((l - l')c_1/(1 + c_1) = l \) is integer and the “interference” term \( 1 - \cos(2\pi l) \) in the numerator vanishes at the same time. These divergences are regularized by taking the finite limit of the quantity \((1 - \cos(e))/\epsilon^2\) when \( \epsilon \to 0 \).

Eq. (4) gives the amplitudes for the \( k \)th harmonics of the combinations \( F = l_0f_0 + l_1f_1 \), \( l_1 \) is a positive integer and \( l_0 \) can be negative. These include “forbidden” frequencies like \( f_1 - f_0 \). The amplitudes depend on whether \( f_0/f_1 \) is equal to an irreducible ratio of 2 integers \( p_1/p_0 \). In this case, we can find many combinations \( l'f_0 + lf_1 \) that are equal to \( kF \). These occur when \( l = kl_1 + mp_1 \) and \( l' = kl_0 - mp_0 \), with \( m \) positive integer. We then obtain

\[
A(kF) = \frac{kF(1 + c_1)^3}{2\pi^3} \times \sum_{m \geq 0} \frac{(-1)^{k(l_0 - l_1) - m(p_0 + p_1) + 1}}{(k(l_1 + c_1 l_0) + m(p_1 - c_1 p_0))^2} \\
\times \frac{1 - \cos \frac{2\pi c_1 (kl_0 - l_1 - m(p_0 + p_1))}{1 + c_1}}{(kl_0 - l_1 - m(p_0 + p_1))^2}.
\]

(5)

In the case where \( f_0/f_1 \) is not rational, only the term \( m = 0 \) has to be taken. We will only consider in the rest of the paper the case when \( f_0/f_1 \) is irrational, for simplification, and therefore only the first term \( m = 0 \) on the right hand side of Eq. (5) contribute. Eq. (5) holds for any frequency except for the special case \( F = k(f_0 + f_1) \):

\[
A(k(f_0 + f_1)) = \frac{c_1}{1 + c_1 \pi k}.
\]

(6)

We obtain a simple result: the \( k \)th harmonics amplitude of \( f_0 + f_1 \) is the same as it would be in GCE case for a single band with fundamental frequency \( f_0 + f_1 \) (except for a \((-1)^k\) sign). The amplitudes for harmonics of \( f_0 \) are:

\[
A(kf_0) = \frac{(-1)^{k+1}(1 + c_1)^3}{2\pi^3 k^3 c_1} \left(1 - \cos \frac{4\pi c_1}{1 + c_1} \right).
\]

The difference with the GCE is that harmonics amplitudes of any frequency but \( f_0 + f_1 \) fall like \( 1/k^3 \) instead of \( 1/k \). We conclude that the jumps in magnetization only come from the contribution of \( f_0 + f_1 \), since its \( k \)th harmonics amplitude decreases like \( 1/k \). The amplitude of the “forbidden” frequency \( f_1 - f_0 \) is:

\[
A(f_1 - f_0) = \frac{(1 + c_1)^3}{(1 - c_1)^2} \left(1 - \cos \frac{4\pi c_1}{1 + c_1} \right).
\]

Figure 1 shows the numerical comparison between GCE and CE amplitudes. Analytical approximation based on thermodynamical relations [8] shows that the magnetization amplitudes \( A(F) \) for combined frequencies \( F = l_0f_0 + l_1f_1 \) at finite temperature are proportional to \( R_{0,l_0}(T)R_{l_1,l_1}(T)F/l_0l_1 \). Basically this is the product of GCE amplitudes for the individual frequencies \( l_0f_0 \) and \( l_1f_1 \). For \( F = kf_0 \), the authors found \( A(F) \propto R_{0,kf_0}. \) At zero temperature the \( k \)th harmonics amplitudes of these frequencies decrease to \( 1/k \) in any case, which is not the exact case since we showed the dependence in \( k \) is \( 1/k^3 \). Moreover, the effect of commensurability between fundamental frequencies is not included. The finite low temperature dependence of the amplitudes is therefore not understood yet. However their approximation may work beyond a crossover temperature below which corrective terms are needed. Indeed the chemical potential fluctuations decrease with increasing temperature and we expect GCE and CE temperature reduction factors be equivalent for frequencies \( kf_0 \).

In conclusion, we computed the exact Fourier magnetization for the 2 band model at zero temperature. The expressions found are different from LK theory except for the harmonics of \( f_0 + f_1 \). The \( k \)th harmonics amplitude of other frequencies decreases like \( 1/k^3 \), and includes an interference term depending on the mass ratio. These results may be useful to check possible low temperature theories.
Fig. 1. Comparison between the absolute amplitudes in the CE (grey line) and GCE (black line) for \( c_1 = m_0^* / m_1^* = 1/2, \Delta_0 = 0.1, \Delta_1 = 0, n_e = 1, f_0 = 4/15 \) and \( f_1 = 11/15 \).

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