Frequency Mixing of Magnetic Oscillations: Beyond Falicov-Stachowiak Theory

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The interpretation of de Haas–van Alphen oscillations in the presence of magnetic breakdown is usually based on the semiclassical theory of Falicov and Stachowiak (FS). There are now glaring discrepancies between its predictions and experiments, especially in quasi-two-dimensional organic conductors. We present an extension of the theory, using the appropriate constraints of conserved electron density, which explains the occurrence of frequencies not predicted by FS, and makes explicit the amplitudes as a function of Fermi surface parameters. The results involve a tunneling amplitude between different sheets as in FS, but other parameters as well, such as the areas of different orbits. [S0031-9007(98)05725-1]

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Magnetic breakdown of oscillations in the magnetization or transport is an important tool in exploring Fermi surfaces. At weak magnetic fields oscillations are visible and correspond to extremal cross sections of semiclassically closed orbits perpendicular to the field direction. If there are several bands there are frequencies corresponding to each closed orbit but not to orbits which traverse the first Brillouin zone and are open. As the magnetic field increases in a system with several bands, there may be magnetic field-induced tunneling from band to band, giving rise to larger orbits with frequencies that are combinations of the original frequencies or which may include parts of open parts of the Fermi surface, giving new frequencies. The characteristic fields at which oscillations appear give information on local properties of the bands: separation of the disjoint parts of the Fermi surface and the local curvature. In the case of bidimensional organic conductors such as the family (BEDT-TTF)$_2X$(SCN)$_4$ where BEDT-TTF is (bis)-ethyleneditho-tetrathiafulvalene [1,2], information on the open parts of the Fermi surface is a key to understanding low temperature instabilities. In addition to extract physical parameters, renormalized masses and g factors in particular, we need a reliable and detailed theory of oscillations. In the past this has been provided by semiclassical theory [3], which culminated in the theory of Falicov and Stachowiak (FS) [4]. In 1982, frequencies were observed [5] for pure magnesium that are, however, forbidden in such a theory. In the organic metals there are more and more violations in the frequency spectrum from experiment. The existence of the "forbidden" β-α frequency (see below) in the magnetoresistance of κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ [6] was attributed to a Stark interference [3,7] but has since been seen clearly in de Haas–van Alphen (dHvA) [8,9], where Stark interference does not apply. Numerical simulation [10–12] showed that the discrepancy is a single particle effect. It has been argued [12–15] that the reason for observations of frequencies corresponding to classically disallowed orbits is that the different frequencies are coupled by the constraint that the total number of electrons across all bands is conserved as the field varies. This global constraint leads to coupling between different frequencies and frequencies forbidden in the FS theory. Such a constraint, without the effects of breakdown, has been shown numerically [14,15] to change the harmonic content of a single frequency. What we lack are explicit calculations of the amplitudes of breakdown when this effect is taken into account. These should be substituted for the FS expressions. In this paper we shall show how FS theory must be amended and we calculate, for a simple case, the correct forms. We also give explicit account of the spin splitting used to extract the renormalized Landé factor g, at least in the low-field limit.

We now consider a simple case of breakdown in two dimensions between open and closed orbits. The Fermi surface, close to that of κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ is shown in Fig. 1. The theory describing the tunneling process at each junction is given in Ref. [16]. We adapt the theory of Ref. [16] to describe the tunneling process at each junction in terms of a gap k_g and curvature $k_g/2\lambda^2$,

$$k_x \pm \sqrt{\lambda^2 k_y^2 + k_g^2/4} = 0\,.$$

If p and q are the amplitudes of tunneling and reflection ($p^2 + q^2 = 1$), and ω the phase the wave function takes during the reflection process, then the transfer relations between the wave amplitudes before and after the junction points are given [16],

$$\begin{pmatrix} \gamma \\ \delta \end{pmatrix} = \begin{pmatrix} q\exp(i\omega) & ip \\ ip & q\exp(-i\omega) \end{pmatrix} \begin{pmatrix} \alpha \\ \beta \end{pmatrix}$$

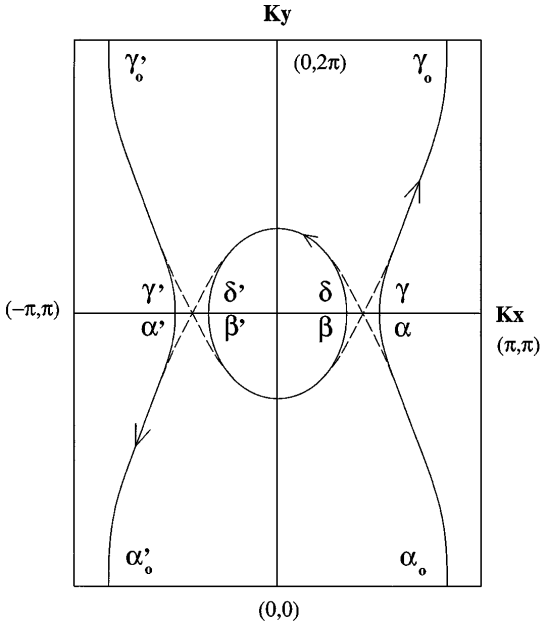

FIG. 1. Typical Fermi surface with one closed pocket at the center of the figure and the two open lines. The magnetic breakdown is represented by the dashed lines.

and similarly for the other junction. The amplitude of tunneling is $p = \exp(-\pi b^2/2\sigma)$, with $b^2 = k_g^2/4\lambda$ and $\sigma = eB/\hbar$. The phase ω is found by matching parabolic cylinder functions,

$$\omega = -\frac{\pi}{4} + \frac{b^2}{2\sigma}\ln\left(\frac{b^2}{2\sigma}\right) - \frac{b^2}{2\sigma} - \arg\Gamma\left(i\,\frac{b^2}{2\sigma}\right).$$

The spectrum is computed with semiclassical methods. Indeed, the phase variation of the wave function from the point δ to δ' is given by $\sigma_2 + \pi/2 \equiv S_2/2\sigma + \pi/2$, where S_2 is the area of the small pocket. The factor $\pi/2$ comes from the existence of a turning point. Similarly, $\alpha = \alpha_0 \exp(i\sigma_1/2)$, where $\sigma_1 = S_1/2\sigma$, S_1 being the area delimited by the two open surfaces. The other relations are similar, and we set the conditions on the edges of the Brillouin zone

$$\gamma_0 = \alpha_0 \exp(i\theta), \qquad \gamma_0' = \alpha_0' \exp(-i\theta)$$

with θ between 0 and 2π. In the following we neglect the phase ω for simplicity since it is small in the semiclassical limit. In fact, it can be absorbed in the new definitions $\sigma_1 + \omega \to \sigma_1$ and $\sigma_2 - \omega \to \sigma_2$. Eliminating the different amplitudes determines the spectrum [16,17]

$$\cos(\sigma_1 + \sigma_2) = 2q\cos\theta\cos\sigma_2 - q^2\cos(\sigma_1 - \sigma_2). \tag{1}$$

The grand thermodynamical potential is determined from the spectrum (1) following the methods of [18,19]. We choose an increasing function of the energy ϵ,

$$2\pi n_\pm(\epsilon,\theta) = \sigma_1 + \sigma_2 \pm [2q\cos\theta\cos\sigma_2 - q^2\cos(\sigma_1 - \sigma_2)].$$

Integer values of n define the spectrum. For small q we recover the Landau quantization of the giant orbit β. In this limit, the argument of the arccosine function vanishes and the variation of n with the energy follows the variation of $\sigma_1 + \sigma_2$, assumed positive near the Fermi energy and for small q. The only restriction is when the arccosine function is no longer defined for all energy and θ, i.e., for $q \geq \sqrt{2} - 1$. In fact, we will check that the final result no longer depends on this restriction. The oscillating part of the potential is

$$\Omega_{\rm osc} = -D\int_0^\pi \frac{d\theta}{\pi}\, 2\sum_{k=1}^{+\infty}\sum_{\sigma=\pm} \times \int_0^{+\infty} d\epsilon f_\beta(\epsilon)\,\frac{1}{2\pi k}\sin(2\pi k n_\sigma),$$

where $f_\beta(\epsilon)$ is the Fermi distribution $1/[1 + \exp\beta(\epsilon - \mu)]$, and $D = g_0 L_x L_y Be/h$ is the degeneracy of each Landau level (g_0 is the spin degeneracy). The usual method at low temperatures is to expand the integrand near the chemical potential (where the Fermi function decreases rapidly), since n_σ is considered large in the semiclassical regime. The oscillatory part of the magnetization is then $M_{\rm osc} = -\partial\Omega_{\rm osc}/\partial B$. Before performing these operations, we sum over σ,

$$\sum_{\sigma=\pm}\sin(2\pi k n_\sigma) = 2\sin k(\sigma_1 + \sigma_2) \times T_k[2q\cos\theta\cos\sigma_2 - q^2\cos(\sigma_1 - \sigma_2)],$$

where the T_k are the Chebyshev polynomials. The integration over θ gives the different frequencies which appear in the Fourier transform of the magnetization. Unfortunately, it is necessary to integrate term by term. For the first terms ($k = 1, 2, 3, \ldots$) we find

$$2\sum_{k=1}^{+\infty}\int_0^\pi \frac{d\theta}{\pi}\sum_{\sigma=\pm}\frac{1}{k}\sin(2\pi k n_\sigma) = q^2\sin(2\sigma_2 + \pi) + (ip)^4\sin(2\sigma_1 + 2\sigma_2 + \pi) + 2(ip)^4 q^2\sin(2\sigma_1 + 4\sigma_2 + 2\pi) + \ldots. \tag{2}$$

The amplitudes appearing with each oscillation are understood as follows: Around the orbit α for example (term $2\sigma_2 = 2\pi F_\alpha/B$), a wave packet is reflected twice at the junctions, and so a factor q^2 is present. For the orbit β ($2\sigma_2 + 2\sigma_1 = 2\pi F_\beta/B$), there are four tunneling junctions, and we put a factor $(ip)^4$, etc. We then add a symmetry

factor for each combination of principal orbits: For $\alpha + \beta$, there are two possibilities to draw such an orbit. At the end, we put a $\pi/2$ factor in the sine functions for each turning point (twice for α or β orbits). These rules are in fact those formulated by FS. We conclude then that working at fixed chemical potential there are no forbidden frequencies present in the Fourier transform of the magnetization: The first terms are α, β, and $\alpha + \beta$. There are no difference frequencies.

We now consider, at zero temperature, the case of the fixed total number of electrons. To simplify, we take a linear relation between the area and the energy,

$$S_i = \frac{2\pi m_i^*}{\hbar^2}\,\epsilon, \qquad i = 1, 2\,.$$

Then we introduce the dimensionless variable x such that $\sigma_1 = 2\pi x$. Finally, the ratio σ_1/σ_2 is fixed to be a rational $r = n/m \geq 1$ with m and n relatively prime. The relation between x and the energy takes the form

$$\epsilon = 2x\hbar\omega_1, \qquad \omega_1 = \frac{eB}{m_1^*}\,,$$

$$\omega_\beta = \frac{n}{m+n}\,\omega_1, \qquad \omega_\alpha = \frac{n}{m}\,\omega_1\,.$$

Periodicity $x \to x + n$ then allows us to work in the sectors $kn \leq x \leq kn + n$, where Eq. (1) has $2(m + n)$ solutions with the symmetry $x \to (2k + 1)n - x$.

We now analyze the expressions of the oscillatory part of the magnetization for both $q = 1$ and q small at fixed electron number. For $q = 1$ (no magnetic breakdown), the Landau levels of the α orbit are embedded in a continuous band. The chemical potential is located either in one Landau band or between two of them. There is no jump in the chemical potential and no jump in the magnetization at zero temperature. The effect of the splitting of the Landau levels due to the electron spin is to reduce the Fourier coefficients of the magnetization. Setting $\Delta = gm_\alpha^*/2m_e$, the oscillatory part of the magnetization per electron is

$$m_{\rm osc} = \frac{\hbar e}{m_\alpha^*}\left[-\sum_{k\geq 1}(-1)^{k(1+[\Delta])}\,\frac{(1+r)^2}{\pi^2 k^2 r}\,2\sin\left[\frac{\pi k}{2(1+r)}\right]\cos\left\{\frac{\pi k}{1+r}\left[(\Delta - [\Delta])r + \frac{1}{2}\right]\right\}\sin(2\pi k F_\alpha/B)\right], \quad (3)$$

where $[\Delta]$ means the integer part of Δ.

It is clear that the amplitudes of the different harmonics depend on the ratio r and are not universal. This is not the case when the chemical potential is fixed, and the different bands can be treated independently. That is why the amplitudes in this case do not then depend on geometrical factors. Finally, we recover the formula for a single band in the limit $r = 0$ with a pure frequency F_α.

We now consider the development for small q (fields much greater than that needed for tunneling). Here we neglect spin. We compute the Fourier transform of the magnetization using an expansion of the energy spectrum at first order in q. In particular, we obtain the weight of the different frequencies appearing in the Fourier spectrum, at least in first order in q. In general, for $q \neq 1$, the amplitudes and phases in the thermodynamical quantities do not follow the simple rules of FS. For small q, the broadening of the β-orbit Landau level is proportional to q. To first order,

$$\epsilon_{s,\sigma} = \epsilon_{s,\sigma}^{(0)} - \sigma\,\frac{2q}{\pi}\,\hbar\omega_\beta\cos(\theta)\cos(\pi\epsilon_{s,\sigma}^{(0)}/\hbar\omega_\alpha)\,,$$
$$\epsilon_{s,\sigma}^{(0)} = d\hbar\omega_\beta\left(2s + \frac{1}{2}\sigma\right), \quad (4)$$

with $\sigma = \pm 1$ labeling, respectively, the even and odd levels. When $\sigma = -1$ the first level begins for $s = 1$, otherwise $s = 0$. After integrating over θ, we find the following energy density:

$$\rho(\epsilon) = \frac{D}{\pi}\sum_{s,\sigma}\frac{\theta(\epsilon - \epsilon_{s,\sigma}^{\rm min})\theta(\epsilon_{s,\sigma}^{\rm max} - \epsilon)}{\sqrt{(2q\hbar\omega_\beta\cos(\sigma_2)/\pi)^2 - (\epsilon - \epsilon_{s,\sigma}^{(0)})^2}}\,, \quad (5)$$

where $\theta(x)$ is 0 or 1, respectively, when x is negative or positive. The width of the levels depends clearly on the value of s and σ, but is linear in q. Finally, we found the oscillatory part of the magnetization,

$$m_{\rm osc} = \frac{\hbar e}{\pi m_\beta^*}\left[\sum_{P\geq 1}\frac{1}{P}\sin(2\pi P F_\beta/B) + q\sum_{P\geq 1}A_P\sin\left(2\pi\,\frac{P}{m+n}\,F_\beta/B\right)\right], \quad (6)$$

with the amplitudes of the correction,

$$A_P = \frac{16P\cos[\pi P/(m+n)]}{\pi[(m+n)^2 - 4P^2]} \times \sum_{k=0}^{m+n-1}\left|\cos\left[\frac{\pi m}{2(m+n)}\,(2k+1)\right]\right| \times \cos\left[\frac{\pi P}{m+n}\,(2k+1)\right]. \quad (7)$$

The frequencies appearing in the correction term are in fact harmonics of $F_1 = F_\beta/(m + n)$. It is useful to express the frequencies appearing in terms of a linear combination of β and α. For an explicit example, we can take $m = 2, n = 7$. The first eleven frequencies correspond to $\beta - 4\alpha$, α, $\beta - 3\alpha$, 2α, $\beta - 2\alpha$, 3α, $\beta - \alpha$, 4α, β, 5α, and $\beta + \alpha$. Between the classical orbits such as β, α, and their harmonics, we recognize other forbidden frequencies which appear simply as harmonics of F_1. The important point is to determine the weight of each amplitude A_P since this determines which will be seen experimentally. We draw in Fig. 2 the histogram of the function

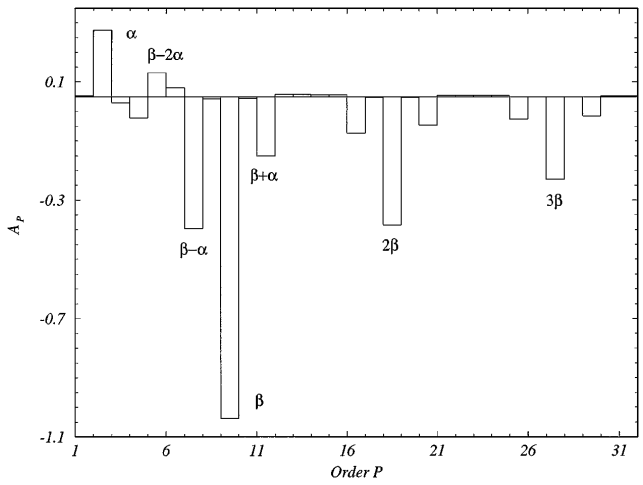

FIG. 2. Histogram of the amplitudes A_P versus P for the case $m = 2, n = 7$.

A_P versus P for the case $m = 2, n = 7$, and in Fig. 3 we plot the semiclassical amplitudes by solving for spectrum (1) numerically.

In conclusion, to include the physical constraint of constant electron density, the FS expressions in Eq. (2) must be replaced by expressions of the form (6) and (7) valid for small q, by (3) for $q = 1$ or more generally by a numerical calculation as in Fig. 3. Unlike the simple FS expressions, the amplitudes depend on the ratio of areas as well as the tunneling probabilities. Extraction of g factors in the limit of low field requires the use of a correct formula as in Eq. (3). The forbidden frequencies occur quite naturally, and their amplitudes can be used to verify the consistency of the Fermi surface parameters.

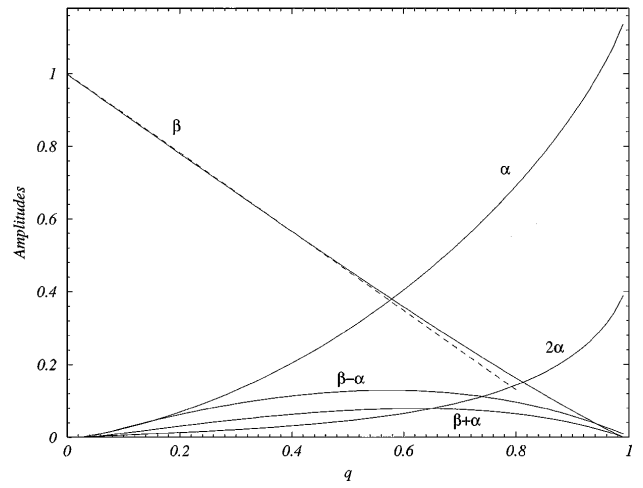

FIG. 3. Amplitudes of different orbits versus q for the case $m = 2, n = 7$ The dashed line is the asymptotic form of the amplitude β calculated for small q

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