Angular dependence of novel magnetic quantum oscillations in a quasi-two-dimensional multiband Fermi liquid with impurities

A.M. Bratkovsky\textsuperscript{1} and A.S. Alexandrov\textsuperscript{2}

\textsuperscript{1} Hewlett-Packard Laboratories, 1501 Page Mill Road, Palo Alto, California 94304
\textsuperscript{2} Department of Physics, Loughborough University, LE11 3TU, United Kingdom

(April 24, 2001)

The semiclassical Lifshitz-Kosevich-type description is given for the angular dependence of quantum oscillations with combination frequencies in a multiband quasi-two-dimensional Fermi liquid with a constant number of electrons. The analytical expressions are found for the Dingle, thermal, spin, and amplitude (Yamaji) reduction factors of the novel combination harmonics, where the latter two strongly oscillate with the direction of the field. At the “magic” angles those factors reduce to the purely two-dimensional expressions given earlier. The combination harmonics are suppressed in the presence of the non-quantized (“background”) states, and they decay exponentially faster with temperature and/or disorder compared to the standard harmonics, providing an additional tool for electronic structure determination. The theory is applied to Sr$_2$RuO$_4$.

The low-dimensional organic conductors exhibit very unusual electronic properties, like magnetic field induced superconductivity, and are the focus of solid state research\textsuperscript{1}. Magnetic oscillations of susceptibility and resistivity are the main source of detailed information about their electronic structure, yet there is no complete theory of these phenomena for quasi-two-dimensional (quasi-2D) metals. The analytical semiclassical (Lifshitz-Kosevich type\textsuperscript{2}) theory of the de Haas-van Alphen combination harmonics in the multiband two-dimensional (2D) metals has been suggested recently\textsuperscript{3}. Even earlier it was shown\textsuperscript{4} for a system like Sr$_2$RuO$_4$\textsuperscript{5}, that the magnetic quantum oscillations in a multiband 2D metal with a fixed electron density [canonical ensemble (CE)] are qualitatively different from those in an open system where the chemical potential is fixed [grand canonical ensemble (GCE)]. The chemical potential oscillates with the field in closed systems and this provides a mechanism for different bands to communicate with each other in the field in closed systems and this provides a mechanism for different bands to communicate with each other in the field in closed systems and this provides a mechanism for different bands to communicate with each other in the field.

Consider the multiband quasi-2D system with a general dispersion law

$$\epsilon = \Delta_\alpha + \frac{k_x^2 + k_y^2}{2m_\alpha} - 2t_\alpha \cos jk_z d, \quad (2)$$

where $k_x, k_y$ are the momenta in the conducting plane, $t_\alpha$ is the hopping between the (conducting) layers, $d$ the distance between the layers, $j$ the integer number. We are mainly interested in the situation where the kinetic energy of electrons is much larger than the hopping between layers, $\mu - \Delta_\alpha \gg 2t_\alpha$. The density of states (DOS) with an account for collision broadening of the Landau levels (Dingle factor)\textsuperscript{6} can be written as

$$\mathcal{N}(\epsilon, B) = \sum_{\alpha,bg} \mathcal{N}_\alpha(\epsilon, B), \quad (3)$$
\[ N_\alpha(\epsilon, B) = \frac{1}{\pi} \text{Im} \sum_{n=0}^{\infty} \frac{\Delta S}{(2\pi)^3} \int_{-\pi/d}^{\pi/d} \frac{dk_z}{\epsilon - \epsilon_{\alpha,nk_z + i\Gamma'_\alpha}}, \]

where \( \Delta S = 2\pi|e|B/\hbar c \) is the cross-sectional area in k-space between the two successive Landau orbits,

\[ \epsilon_{\alpha,nk_z} = \Delta_{\alpha 0} + \omega_\alpha (n + 1/2) + D_\alpha \cos jk_z d + g_\alpha \sigma_B \mu_B B, \]

the energy dispersion for a present geometry of the Landau orbits, where \( D_\alpha = 2t_\alpha J_0(jk_\alpha d \tan \theta) \) with \( J_0(x) \) the zeroth-order Bessel function, \( \Gamma'_\alpha = \pi/2\tau_\alpha, \tau_\alpha \) the scattering mean free time in zero field \([4]\) in the band \( \alpha \), and the background \((bg)\) is included in the DOS \([3]\) too \([3]\). After integration we obtain an important exact result

\[ N_\alpha(\epsilon, B) = \frac{1}{\pi} \text{Im} \sum_{n=0}^{\infty} \rho_\alpha \omega_\alpha \nu(\epsilon - \epsilon_{\alpha,n}), \]

\[ \nu(\epsilon - \epsilon_{\alpha,n}) = \text{Im} \left( \frac{i}{\pi \sqrt{D^2_\alpha - (\epsilon - \epsilon_{\alpha,n} + i\Gamma'_\alpha)^2}} \right), \]

where \( k^2_\alpha / 2m_\alpha = \mu - \Delta_\alpha \),

\[ \epsilon_{\alpha,n} = \Delta_{\alpha 0} + \omega_\alpha (n + 1/2) + g_\alpha \sigma_B \mu_B B, \]

\( \omega_\alpha = |B \cos \theta/m_\alpha c | \) the cyclotron frequency with the cyclotron mass \( m_\alpha \), \( \Delta_{\alpha 0} \) the band edge in zero magnetic field, \( \mu \) the chemical potential, \( g_\alpha \) the electron g-factor, \( \sigma = \pm 1/2 \), \( \mu_B \) the Bohr magneton. The band index \( \alpha = bg \) includes the band index b and the spin index \( \sigma \). There \( \rho_\alpha \) is the zero-field density of states in the band \( \alpha \). For the energies of interest, \(|\epsilon - \Delta_\alpha| > 2t_\alpha \), \( \rho_\alpha = m_\alpha / 2\pi \hbar^2 d \) per energy and unit volume, otherwise \( \rho_\alpha = (m_\alpha / 2\pi^2 \hbar^2 d) \arccos[(\epsilon - \Delta_{\alpha 0})/2t_\alpha] \). Note that in a clean system at the magic angles, where \( D_\alpha = \Gamma'_\alpha = 0 \), the expression for \( \nu \) reduces to \( \nu(\epsilon - \epsilon_{\alpha,n}) = \delta(\epsilon - \epsilon_{\alpha,n}) \), meaning that the spectrum becomes 2D, the Landau minibands reduce to the Landau levels, and the previous expressions \([3]\) fully apply. The background density of states, \( \rho_{bg} \), corresponding to possible non-quantized (or largely broadened) bands, can be included in \([3]\) as the formal limit \( \omega_{bg} \rightarrow 0 \). Obviously, the non-quantized background will contribute to the non-oscillating characteristics of the system. In a clean limit the density of states takes a standard form with the one-dimensional square-root singularities \( \mathcal{N} \sim 1/\sqrt{D^2_\alpha - (\epsilon - \epsilon_{\alpha,n})^2} \) \([4]\).

By applying the Poisson formula \([2]\) to the sum over \( n \) in the thermodynamic potential

\[ \Omega = -T \sum_{n=0}^{\infty} \int_0^\infty \frac{d\epsilon}{2\pi} \rho_\alpha \omega_\alpha \nu(\epsilon - \epsilon_{\alpha,n}) \]

\[ \times \ln \left( 1 + e^{\mu - \epsilon/T} \right) \]

with \( \mu_\alpha = \mu - \Delta_\alpha \) and \( \Delta_\alpha = \Delta_{\alpha 0} + g_\alpha \sigma_B \mu_B B \), it can be written as

\[ \Omega = \Omega_0 + \tilde{\Omega}, \]

where, after substituting \( x = (\epsilon - \epsilon_{\alpha,n})/D_\alpha \),

\[ \Omega_0 = -T \sum_{\alpha,bg} \int_0^\infty \frac{d\epsilon}{2\pi} \rho_\alpha \sum_{n=1}^{\infty} \int_{-\infty}^\infty \frac{dx}{\pi} \text{Im} \left( \frac{i}{\sqrt{1 - (x + i\Gamma'_\alpha)^2}} \right) \]

\[ \times \ln \left( 1 + e^{\mu_\alpha - \epsilon - D_\alpha x/T} \right) \]

is the “classical” part of the thermodynamic potential, with \( \Gamma'_\alpha = \Gamma'/D_\alpha \). In the GCE \( \Omega_0 \) does not oscillate as a function of \( 1/B \), and contains the contribution due to spin susceptibility (Pauli paramagnetism). At low temperatures one finds

\[ \Omega_0 = -\sum_{\alpha,bg} \frac{1}{\pi} \rho_\alpha \left( \mu_\alpha^2 + \frac{1}{2}D_\alpha^2 \right). \]

The oscillating part of the thermodynamic potential includes only the Landau quantized bands,

\[ \tilde{\Omega} = -2T \sum_{\alpha,bg} \sum_{n=1}^{\infty} \int_0^\infty \frac{d\epsilon}{2\pi} \int_{-\infty}^\infty \frac{dx}{\pi} \text{Im} \left( \frac{i}{\sqrt{1 - (x + i\Gamma'_\alpha)^2}} \right) \]

\[ \times \ln \left( 1 + e^{\mu_\alpha - \epsilon - D_\alpha x/T} \right) \cos 2\pi r \left( \frac{\epsilon}{\omega_\alpha} - \frac{1}{2} \right). \]

This expression, after integrating over \( \epsilon \) and \( x \), reduces to

\[ \tilde{\Omega} = \frac{1}{24} \sum_{\alpha} \rho_\alpha \omega_\alpha^2 + 2 \sum_{\alpha} \sum_{r=1}^{\infty} A^r_\alpha \cos 2\pi r \left( \frac{F_\alpha}{B} - \frac{1}{2} - \psi_\alpha \right) \]

\[ = \frac{1}{24} \sum_{\alpha} \rho_\alpha \omega_\alpha^2 + 4 \sum_{b} \sum_{r=1}^{\infty} A^r_b \cos 2\pi r \left( \frac{F_b}{B} - \frac{1}{2} \right), \]

where \( F_b = (\mu - \Delta_{\alpha 0})|m_b|/2\mu_B m_\alpha c \equiv \hbar c S_{fb}/2\pi e, \)

\( S_{fb} \) is the mean Fermi surface zero-field cross-section (in standard units), and the spin-related phase \( \psi_\alpha = \sigma^b \gamma_b = g_b |m_b|/(2m_\alpha c \cos \theta) \). If the scattering time were not dependent on the spin projection, the phase would be the only quantity explicitly depending on spin in Eq. \([14]\), hence one can perform a summation over \( \sigma \) in the second term to reveal the standard spin reduction factor \( \cos \pi r \gamma_b \).

The amplitudes of the Fourier harmonics in Eq. \([14]\) are explicitly given by

\[ A^r_b = \frac{\rho_b \omega_b^2}{4\pi^2 r^2} R_T \left( \frac{2\pi^2 r T}{\omega_b} \right) R_Y \left( \frac{2\pi R D_b \omega_b}{\omega_b} \right) R_D R_s, \]

where \( R_T(z) = z/\sinh z \) is the usual temperature reduction factor \([4]\), \( R_Y(z) = J_0(z) \) the orientation (Yamaji) factor noticed in \([3]\), \( R_D = e^{-2\pi r \Gamma_b / \omega_b} \) the Dingle exponential damping due to collision broadening of the Landau minibands, \( R_s = \cos \pi r \gamma_b \) the spin reduction factor, which all strongly depend on the orientation of the field,
angle θ (Fig. 1). As usual, one can view the Dingle factor as corresponding to the effective temperature $T + T_D$ in a clean system, where the Dingle temperature $T_D = \Gamma_b/\pi$. For the magic angles, where $D_\alpha = 0$, one has $R_Y = 1$ and hence a pure 2D situation is recovered [3]. However, generally in a multiband case one should expect that only a spectrum of a particular band will become 2D at some particular magnetic field tilt angle.

The expression for $\tilde{\Omega}$, Eq. (14), contains the (first) term, responsible for the Landau diamagnetism and the oscillating term, which is responsible for the de Haas-van Alphen effect. It is small compared with the “classical” part, since $\tilde{\Omega}/\Omega_0 \sim (\omega/\mu)^2 \ll 1$ for the present semiclassical situation. The Fourier components appear with the frequencies $rF_\alpha$. We recover from $\Omega_0$ [12] and the first term in $\tilde{\Omega}$ [13] the Pauli $\chi_P = \frac{1}{2} \mu_B^2 \sum_\alpha \rho_\alpha g_\alpha^2$ and the Landau $\chi_L = -\frac{1}{3} \mu_B^2 \cos^2 \theta \sum_\alpha \rho_\alpha (m_\alpha/m_e)^2$ susceptibilities, respectively. Since usually $g_\alpha = 2$, those are almost standard, apart from the $\cos^2 \theta$ factor characteristic of the present geometry with the tilted magnetic field.

Note that the chemical potential (and carrier density) oscillates in a closed system and, unlike in GCE, the “classical” part of $\Omega$ contributes to oscillations as well [3]. The relevant thermodynamic potential of the closed system (CE) is the free energy, $F = \Omega + \mu N$, for a fixed number of electrons, $N = -\partial \Omega/\partial \mu$. The chemical potential is

$$\mu = \mu_0 + \tilde{\mu},$$

$$\tilde{\mu} = -\frac{\tilde{N}}{\rho} = -\frac{\tilde{N}}{\rho_\alpha + \rho_{bg}}$$

where $\mu_0$ is the non-oscillating, while $\tilde{\mu}$ is the oscillating part of the chemical potential, $\tilde{N} = -\partial \tilde{\Omega}/\partial \mu$ is the oscillating part of the density of electrons, $\rho = \sum_\alpha \rho_\alpha \equiv \rho_\alpha + \rho_{bg}$ is the total density of states, including the quantized $\rho_\alpha$ and the background $\rho_{bg}$ DOS. Substituting this expression into $\Omega_0$, Eq. (13), we obtain $F = F_0 + \tilde{F}$, where the oscillating part is

$$\tilde{F} = \tilde{\Omega} - \frac{\tilde{N}_\alpha}{2\rho_\alpha},$$

while $F_0$ is the non-oscillating part. One sees that the difference between the free energies of the ensembles is directly proportional to the fluctuation of the particle density, as it should, and is suppressed by the background density of states $\rho_{bg}$, $\sim N^2/(\rho_\alpha + \rho_{bg})$. The oscillating part of the particle density is small, but both terms in (18) give comparable contribution to the magnetization oscillations. Indeed, $\tilde{N} = \sum_\alpha \tilde{N}_\alpha$, and at low temperatures

$$\frac{\tilde{N}_\alpha}{N_0} \sim \frac{\rho_\alpha B}{\rho F_\alpha} \ll 1, \quad T < \omega_\alpha,$$

since $F_\alpha \gg B$, and it falls off exponentially with temperature, $\tilde{N}_\alpha/N_0 \sim (T/\mu) \exp\left(-2\pi^2 T/\omega_\alpha\right)$, at $T > \omega_\alpha$. Since the density oscillations are small, sometimes they can be screened by the back electrode in e.g. quantum well structures [13]. In a more explicit form one obtains

$$\tilde{F} = \frac{1}{24} \sum_\alpha \rho_\alpha \omega_\alpha^2 + 4 \sum_{b,b'} \sum_{r=1}^{\infty} A_b^r \cos 2\pi r \left(\frac{F_b}{B} - \frac{1}{2}\right)$$

$$-16 \sum_{b,b',r,r'} \sum_{\infty} C_{bb'}^{rr'} \sin 2\pi r \left(\frac{F_b}{B} - \frac{1}{2}\right) \sin 2\pi r' \left(\frac{F_{b'}}{B} - \frac{1}{2}\right).$$

It is the last term, which yields the combination Fourier harmonics with the frequencies $F = rF_b \pm r'F_{b'}$. Their amplitudes,

$$C_{bb'}^{rr'} = \frac{2\pi^2 r r' A_b^r A_{b'}^{r'}}{(\rho_\alpha + \rho_{bg}) \omega_\alpha \omega_{b'}}$$

are comparable with the standard single-band harmonics at low temperatures, $T < \omega_\alpha/2\pi^2 r$, as found earlier [3] and confirmed experimentally [13]. The combination harmonics are suppressed in presence of the background density of states. The spin factor depends on the tilt angle, and this results in a strong angular dependence of the corresponding dHvA amplitudes [13,14]. Incidentally,
if one of the angular dependent factors vanishes for some particular harmonic, \( r_0 F_b \), it would not mix up with other bands to produce combination harmonics, whereas \( r F_b \) \((r \neq r_0)\) would (cf. numerical results [17]).

Similar to the usual 3D situation, the oscillations produce the (partial) contributions to magnetization, \( M_\alpha \), and susceptibility, \( \chi_\alpha \), which are much larger than the non-oscillating contributions. Indeed, the ratio of corresponding amplitudes at low temperatures is \(|\chi^c_\alpha|/\chi_0 \sim (F_\alpha/B)^2 (\rho_\alpha m/\pi^2 \rho m_0 |rJ_0(2\pi rD_\alpha/\omega_\alpha) e^{-2\pi rD_\alpha/\omega_\alpha} r_\alpha/\omega_\alpha \gg 1\), since \( F_\alpha/B \gg 1 \). Proportionality to \((F_\alpha/B)^2\) is the property of the two-dimensional geometry [3]. The amplitudes of the standard and the combination harmonics can be easily found from the expressions given above.

The ratio of susceptibilities is

\[
N^c_{F_b} = \frac{\chi^c_{r F_b \pm r' F_b}}{\chi^c_{r F_b}} = \frac{8\pi^2 r' F_{b'}}{\rho_\alpha m_0 \omega_\beta} \cos \pi r' \gamma b' \left( \frac{F_b}{r F_b} \right)^2,
\]

with \( F = r F_b \pm r' F_{b'} \). Thus, for \( r = r' = 1 \) we have, for a warped 2-band cylindrical Fermi surface without the background DOS,

\[
\chi^c_{F_b \pm F_{b'}}/\chi^c_{F_b} = 4\pi^2 m_\beta T J_0(2\pi D_\beta/\omega_\beta) \cos \gamma b' e^{-2\pi r D_\beta/\omega_\beta} \sinh (2\pi r T/\omega_\beta) \left( \frac{F_b \pm F_{b'}}{F_b} \right)^2,
\]

where \( \chi^c_\beta \) denotes the amplitude of the standard dHvA harmonic for an open system, with \( \chi^c \) the amplitude of the novel combination harmonics. We see that the combination harmonics are suppressed exponentially with respect to temperature [3] and/or disorder compared to the standard harmonics, Fig. 2. In addition, they contain the product of the angular dependent reduction factors and, therefore, vary rapidly with the angle \( \theta \), their frequency being dominated by the band with the largest ratio \( 2\theta_\gamma/\omega_\beta \) (e.g. \( \beta^- \)-band in \( \beta \pm \alpha \) harmonics in \( \text{Sr}_2\text{RuO}_4 \), Fig. 1).

It is important to note that even in a simplest case of a one-band system the standard harmonics \( r F_b \) are modified by the second quadratic term, therefore

\[
N^c_{F_b} = \chi^c_{r F_b} = \sum_{p=1}^{r-1} \chi_{r(\mu-p) F_b, p F_b}^c + \sum_{p=1}^{\infty} \chi_{r(\mu+p) F_b, -p F_b}^c = \sum_{p=1}^{\infty} \chi_{r(\mu+p) F_b, -p F_b}^c,
\]

and the wavefront of magnetization of the main harmonics in the closed system with the carrier density \( N = \text{const} \) is inverted compared to the open system, \( \mu = \text{const} \), as it should [4,11].

The present formalism is applied to \( \text{Sr}_2\text{RuO}_4 \) with the results shown in Figs. 1 and 2, calculated with the parameters from Refs [10,13] and the g-factor \( g_b = 2 \) for all the bands. The main maxima in the Fourier harmonics of susceptibility \( \chi \) for the band \( \alpha \) are found at \( \theta_\alpha(2) = 33^\circ \) \((55^\circ), \) for the band \( \beta \) at \( \theta_\beta \approx \theta_\alpha, \) and for band \( \gamma \) at \( \theta_\gamma(1) = 27^\circ \) \((49^\circ), \) which is in fair agreement with the data \( \theta_\alpha = 30.6^\circ, \theta_\beta = 30^\circ \) and \( \gamma = 15.3^\circ, \) Ref. [11]. The g-band, and \( \theta_\alpha = 30^\circ, \theta_\beta(2) = 26^\circ \) \((56^\circ), \) Ref. [1]. Those maxima in the harmonics apparently coincide with the points where the Yamaji factor is unity, \( R_Y = 1, \) and the system effectively becomes 2D, Figs. 1(b), (d). The minima, observed in \( \alpha \) band at 24° [10,25,26] and 40°, are very close to the points were both the Yamaji \( R_Y \) and the spin \( R_s \) factors vanish: \( R_Y = 0 \) at 23° and 41°, and \( R_s = 0 \) at 21° and 43°, Fig. 1(d).

The conspicuous rapid variation with the field angle \( \theta \) of the main, \( \chi_{\beta}, \chi_{\gamma}, \) and the combination, \( \chi_{\beta^- \alpha}, \) harmonics, Fig. 1(a), is explained by the large factor \( 2t_\beta/\omega_\gamma \) in the argument of the Bessel function in the Yamaji factor, Eqs. [15]. Those rapid variations with angle \( \theta \) might be observable, since the condition for the Yamaji approximation, \( \pi k_{f_b}^2 (t_\beta/\epsilon f)^2 \ll \Delta S \) seems to hold for the \( \beta^- \)-band \( [\pi k_{f_b}^2 (t_\beta/\epsilon f)^2 : \Delta S \approx 1 : 5], \) while for the \( \gamma \)-band the ratio is only about 1 : 3. Interestingly, the spin-factor \( R_s \) defines the envelope of the \( \beta^- \)-amplitude, cf. Figs. 1(a) and 1(b), so the studies of the minima and maxima on the \( \chi (\theta) \) should allow for accurate determination of the g-factors \( g_b \).

The combination harmonics contain the extra temperature and the Dingle reduction factors, so they are falling off with either temperature or disorder, or both, faster than the standard harmonics do, see Eqs. (21), (22), Fig. 2.
The temperature dependence of the combination harmonics generally cannot be characterized by some effective mass $m_{rb,\pm r'b'}$, although at higher temperatures the relation $m_{rb,\pm r'b'} \approx rm_b + r'm_b'$ holds approximately. All the harmonics decay quickly with $T$, especially in the presence of even weak disorder (the Dingle temperature $T_D = 0.63K$ [10]), Fig. 2(b), which is in very good agreement with experiment (cf. Fig. 2 in Ref. [10]).

Importantly, in the first experiments on Sr$_2$RuO$_4$ the disorder was actually larger, since the elastic mean free path was $l \sim 10^3\text{Å}$ [5], compared to $l \sim 2050-5000\text{Å}$ in the later experiments [10,11]. The higher disorder in the first samples, and relatively small magnitude of the combination peaks at the field $B$ directed along the c-axis, $\theta = 0$, instead of the ”magic” angle $\theta = 30^\circ$, where all the amplitudes are enhanced, Fig. 1, has possibly prevented the discovery of the combination harmonics, predicted in Ref. [4], in the first experiments on Sr$_2$RuO$_4$ [5]. The combination harmonics have been detected in Refs. [9–11]. The dependence of the dHvA combination amplitudes on angle, temperature, and disorder provide additional valuable tool for studying the band structure and carrier densities in the multiband quasi-two-dimensional metals.

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