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Comment on “Origin of combination frequencies in quantum magnetic oscillations of two-dimensional multiband metals”

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We analyze the applicability of our analytical theory of combination harmonics in canonical low-dimensional multi-band Fermi liquids, which was recently criticized by T. Champel [Phys. Rev. B **65**, 153403 (2002)]. It is shown that his claim that our analytical theory does not apply at low temperatures and in clean samples is incorrect. We demonstrate that the analytical theory of combination harmonics is in excellent agreement with the exact numerical results even at zero temperature and for clean systems, which are the most challenging for an analytical description.

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The combination frequencies in the magnetization of the two-dimensional (2D) multiband metals with constant net electron density have been predicted, given simple physical explanation, and described numerically in Ref. 1. They were further numerically studied in Refs. 2 and 3 and observed experimentally by Shepherd *et al.*⁴ and by other groups.⁵ We have also proposed an analytical theory⁶ of the combination Fourier components in the framework of the semiclassical Lifshitz-Kosevich approach.⁷ More recently the theory has been extended by taking into account the Dingle, spin and angle (Yamaji) reduction factors, and the nonquantized “background” density of states in quasi-2D multiband metals.⁸ There are strong oscillations of a chemical potential pinned to the peaks in the Landau density of states in (quasi) 2D multiband metals, that cause this effect.¹ With the increase of the dimensionality⁸ or background density of states,² the oscillations are strongly damped and the effect vanishes. Smallness of the chemical potential oscillations in 3D metals was already discussed by Dingle in 1951. The purpose of the present paper is to analyze our analytical theory in view of critical remarks in Ref. 9, who claimed that “the chemical potential oscillations appearing in the arguments of the Fourier components were not taken into account” by the present authors. We attempt to clarify the relevant issues pertaining to our analytical expressions, in order to indicate explicitly the approximations made in the derivations and resolve problems with their interpretation. Importantly, we also demonstrate that the analytical results for amplitudes of combination harmonics are numerically accurate even in most unfavorable circumstances at zero temperature and for clean samples.

The basic equations of the theory⁶ are those for the oscillating part $\tilde{F}$ of the free energy, which is the thermodynamic potential of the canonical ensemble,

$$\tilde{F} = \tilde{\Omega}(\mu) - (2\rho)^{-1}(\partial\tilde{\Omega}/\partial\mu)^2, \quad (1)$$

and for the chemical potential $\mu = \mu_0 + \tilde{\mu}$, where $\tilde{\mu} = \rho^{-1}\partial\tilde{\Omega}/\partial\mu$ [Eq. (12) and Eq. (9) of Ref. 6, respectively]. Here $\mu_0 = \rho^{-1}(N + \sum_{\alpha}\rho_{\alpha}\Delta_{\alpha})$ is the zero-field chemical potential, ρ and ρ_{α} are total and partial zero-field densities of

states (DOS), and Δ_{α} is the α -band edge. The oscillating part of the grand canonical potential $\tilde{\Omega}$ [Eq. (6) of Ref. 6] is given by the standard expression¹⁰

$$\tilde{\Omega}(\mu) = 2 \sum_{\alpha} \sum_{r=1}^{\infty} (-1)^r A_{\alpha}^r \cos\left(\frac{rf_{\alpha}}{B}\right), \quad (2)$$

where A_{α}^r are the amplitudes of the single-band Fourier harmonics. For the sake of simplicity, we take the spin-splitting g factors to be zero. Substituting this expression into $\tilde{F}$, one obtains the combination amplitudes $C_{\alpha\alpha'}^{rr'}$ as⁶

$$C_{\alpha\alpha'}^{rr'} = 2\pi^2 \frac{rr' A_{\alpha}^r A_{\alpha'}^{r'}}{\rho \omega_{\alpha} \omega_{\alpha'}}, \quad (3)$$

where $\omega_{\alpha} = eB/m_{\alpha}$. Our definition of the frequencies $f_{\alpha} = 2\pi m_{\alpha}(\mu - \Delta_{\alpha})/e$ in Eq. (2) and in Ref. 6 contains the *exact* chemical potential μ rather than its zero-field value μ_0 . Hence, Eqs. (12) and (13) of Ref. 6 are *exact*. They fully take into account the chemical potential oscillations in the arguments of the Fourier components.

Certainly, we did not consider our explicit expression for $\tilde{F}$, Eqs. (12) and (13) (Ref. 6) as a final Fourier series, as should be obvious from our using the exact μ , not its smooth part μ_0 , in the expression for the free energy. The free energy $\tilde{F}$ has been expanded in powers of $\tilde{\mu} \ll \mu_0$ as

$$\tilde{F} \approx \tilde{\Omega}(\mu_0) + (2\rho)^{-1}(\partial\tilde{\Omega}(\mu_0)/\partial\mu_0)^2, \quad (4)$$

and, differentiating it with respect to magnetic field, we have obtained the result, Eq. (16),⁶ for the magnetization amplitudes. The ratio of the combination $M_{\alpha\alpha'}^{11}$, and single band (conventional) M_{α}^1 Fourier amplitudes in a two-band metal was found at $T=0$ to be [Eq. (16) in Ref. 6]

$$\frac{M_{\alpha\alpha'}^{11}}{M_{\alpha}^1} = \frac{m_{\alpha}}{m_{\alpha} + m_{\alpha'}} \frac{f_{\alpha} \pm f_{\alpha'}}{f_{\alpha}}. \quad (5)$$

It was generalized in Eqs. (21) and (22) of Ref. 8 by taking into account a background DOS, the Dingle, spin, and

FIG. 1. (Color online) Relative ratio of the first approximation M^11_{first} , Eq. (9), and exact M^11 combination amplitudes for a range of Dingle factors R_D . Note that the difference is in the range of 1% or less for both dirty and clean systems. We have assumed $f_2/f_1 = 7/11$. Inset: the oscillating part of the chemical potential $z = 2\pi\tilde{\mu}/\omega$ as a function of inverse magnetic field $1/B$. The two curves correspond to clean (the Dingle factor $R_D = 0.99$, large amplitude of chemical potential oscillations $2\pi\tilde{\mu}/\omega$) and dirty ($R_D = 0.1$, small amplitude) samples, respectively.

Yamaji reduction factors. Obviously, the same result for the magnitude of this ratio can be obtained by differentiating our exact free energy, Eqs. (12) and (13) in Ref. 6, with respect to the magnetic field B but keeping frequency f_α constant. Therefore, the coefficients $C^{rr'}_{\alpha\alpha'}$ in the second term of the free energy indeed yield the correct combination frequency amplitudes in the magnetization, as we pointed in our original paper.⁶ The expansion of the free energy in powers of $\tilde{\mu}$ described above is straightforward, and it was not mentioned explicitly in Ref. 6. The present discussion should clarify the point that the combination frequencies are fully defined by the second-order expansion coefficients $C^{rr'}_{\alpha\alpha'}$.

Alternatively one can first differentiate the free energy, Eq. (1) with respect to B as,¹

$$\frac{\partial \tilde{F}}{\partial B} = \left(\frac{\partial \tilde{\Omega}}{\partial B} \right)_\mu + \frac{\partial \tilde{\Omega}}{\partial \mu} \frac{\partial \tilde{\mu}}{\partial B} - \frac{1}{\rho} \frac{\partial \tilde{\Omega}}{\partial \mu} \frac{\partial}{\partial B} \frac{\partial \tilde{\Omega}}{\partial \mu} = \left(\frac{\partial \tilde{\Omega}}{\partial B} \right)_\mu, \quad (6)$$

which gives the exact expression for the magnetization [Eq. (5) of our original paper¹]. Then one can expand the result in powers of $\tilde{\mu}$, as it was done by Champel in the second part of his paper.⁹ Because the derivatives with respect to μ and with respect to B commute, the approximate amplitudes should be the same as in Eq. (5). Indeed, the exact derivation of the magnetization, the approximate Fourier amplitudes, and the “main” result, Eq. (14) of Ref. 9, are identical to our original expressions, Eq. (5) of Ref. 1 and Eq. (16),⁶ Eqs. (21) and (22),⁸ respectively.

Champel mentions that “... the mechanism responsible for the combination frequencies in the Fourier spectrum of magnetization oscillations cannot apriori depend on the way the magnetization is derived, that is to say, on the use of a specific thermodynamic potential. Our following goal is then

to point out how the combination frequencies arise by considering directly the expression for the magnetization oscillations in the relevant thermodynamic limit.” Indeed, our results do not depend on the use of a particular thermodynamic potential. The mechanism of novel combination frequencies¹ is based on the effect of chemical potential oscillations, which is important in quasi-2D closed metallic systems and is absent in open systems. It can be derived with the use of either thermodynamic potential, its particular selection being a matter of convenience. To imply otherwise would be to misrepresent our work. Obviously, a natural choice of a thermodynamic potential for an open system (with a constant chemical potential) is $\Omega(\mu)$, while for a closed system with the constant number of particles N this would be $F(N)$. One is free to use $\Omega(\mu)$ at constant N as far as one accounts for the functional relation $\mu = \mu(N)$ in the derivation, although it only makes the derivation cumbersome without changing any results. The author of Ref. 9 seems to realize this, in fact using our formulas, e.g., Eqs. (3) and (4) in Ref. 9, to rederive our prior results e.g., Eq. (14) in Ref. 9.

The difference between the corresponding free energies F and Ω of the measured system is tiny, since it is proportional to the fluctuation of the carrier density, Eq. (1). However, the effect on susceptibility is greatly amplified, since two differentiations with respect to the field bring about very large factor $(f/B)^2 \gg 1$, Ref. 8, similar to the case of the usual de Haas–van Alphen effect. Obviously, there will be no chemical potential oscillations when it is fixed by a reservoir, so that no combination frequencies due to these oscillations can be observed in an open (grand canonical) system. The reservoir commonly implies a large system with an infinite continuum of nonquantized electron states. In this sense the role of the reservoir may be played by, e.g., a one-dimensional (and, therefore, not closed) electron orbit with very large “background” density of states in the same quasi-2D sample,² observed in some cases. On the contrary, the unusual combination frequencies appear when the system is closed, so that the number of carriers stays the same, and the chemical potential must oscillate.¹ In actual dHvA experiments the sample is usually measured while placed on nonconducting substrate with no electrodes attached, so the system is indeed closed. Note that the definition of the chemical potential does *not* require that the system be open. For normal Fermi liquids with the ground-state energy $E_0(k)$ for system of k particles the chemical potential $\mu = E_0(k+1) - E_0(k)$ is uniquely defined for large k .

Further, Champel writes about the origin of combination frequencies: “... in 2D metals, the magnetization oscillations become more or less sensitive to the presence of the chemical-potential oscillations and exhibit significantly different behaviors depending on the presence or absence of a finite reservoir of electrons.^{2,9,10} In 2D multiband metals this higher sensitivity to chemical-potential oscillations is expressed by the presence of the combination frequencies in the Fourier spectrum of magnetization oscillations at very low temperatures, as shown numerically by Nakano¹¹. Here, our aim is to prove analytically the existence of these combination frequencies in the magnetization oscillations.” One would note that the first claim that the dHvA oscillations

have qualitatively different wave front in 2D one-band metal compared to 3D case is well known for single-band 2D metals since the pioneering work by Peierls in 1933.¹¹ One would easily see that the existence of combination frequencies has been proven numerically in our work¹ prior to Nakano's work. The analytical proof of their existence has already been given in Refs. 6 and 8.

Now we would like to address the claim of Ref. 9 that “the quantitative description of dHvA oscillations in terms of a Fourier series may break down and has to be done numerically.” Champel⁹ has failed to mention that the numerical description of combination harmonics was done in our original¹ and subsequent papers.^{2,8} To illustrate the accuracy of the analytical theory,^{6,8} let us analyze a simple two-band model. The results below show explicitly that the higher powers of $\tilde{\mu}$ expansion can be neglected even at zero temperature and in a clean sample. Consider two bands with equal masses $m_1=m_2\equiv m$ and equal Dingle reduction factors $R_D=\exp(-2\pi\Gamma/\omega)$, but with the different dHvA frequencies f_1 and f_2 . This two-band model is the most unfavorable case for the first-order expansion because in the presence of more bands the oscillations of the chemical potential are reduced compared to the two-band case. The derivative of $\tilde{\Omega}$, Eq. (2), with respect to μ and the derivative of $\tilde{F}$, Eq. (6), with respect to B yield for $T=0$,

$$z = - \sum_{r=1}^{\infty} \frac{(-R_D)^r}{r} [\sin(rz + rf_1/B) + \sin(rz + rf_2/B)], \quad (7)$$

and

$$\tilde{M} = \frac{e^2}{4\pi^3 m} \sum_{r=1}^{\infty} \frac{(-R_D)^r}{r} [f_1 \sin(rz + rf_1/B) + f_2 \sin(rz + rf_2/B)], \quad (8)$$

where $z \equiv 2\pi\tilde{\mu}/\omega$, and $\tilde{M}$ is the oscillating part of the magnetization ($\omega = eB/m$). Expanding $\tilde{M}$ in powers of z and neglecting z in the right-hand side of Eq. (7) yields the amplitudes of the $(f_1 \pm f_2)$ Fourier components as⁶

$$M_{first}^{11} = \mp \frac{e^2 R_D^2}{16\pi^3 m} (f_1 \pm f_2). \quad (9)$$

One can estimate the accuracy of this expression by Fourier transforming Eq. (8) without the expansion but leaving only

the leading ($r=1$) harmonics in the chemical potential, $z \approx R_D[\sin(f_1/B) + \sin(f_2/B)]$. Then the ratio of the “exact” combination amplitude to the approximate first-order amplitude is given by

$$\frac{M^{11}}{M_{first}^{11}} \approx \frac{2J_0(R_D)J_1(R_D)}{R_D}, \quad (10)$$

where $J_n(x)$ is the Bessel function. This ratio is very close to unity at any value of the Dingle factor. In fact, the first-order expansion⁶ has even better accuracy than the estimate given by Eq. (10).

For the numerical calculations we have used the analytical expressions of the sums in Eqs. (7) and (8),

$$z = \text{Arg}\{[1 + R_De^{i(z+f_1/B)}][1 + R_De^{i(z+f_2/B)}]\}, \quad (11)$$

and

$$\tilde{M} = - \frac{e^2 f_1}{4\pi^3 m} \text{Arg}\{[1 + R_De^{i(z+f_1/B)}] \times [1 + R_De^{i(z+f_2/B)}]^{f_2/f_1}\}, \quad (12)$$

where $\text{Arg}w$ stands for the argument of the complex number w . The oscillating part of the chemical potential and the relative difference between the analytical and numerical amplitudes are shown in Fig. 1. The first-order Fourier amplitudes of the magnetization are equal to the numerical amplitudes within a few percent at any value of the collision broadening Γ . According to our theory, higher combination harmonics, like $f_1 + 2f_2$, should be exponentially suppressed compared to the leading harmonics $f_1 \pm f_2$ studied here. It is not surprising, therefore, that the higher combination harmonics have not been clearly resolved even in very clean samples.¹² Evidently, the present results for the combination $f_1 \pm f_2$ harmonics are sufficient to refute Champel's general claim that our analytical theory loses accuracy at low temperatures in clean systems. Indeed, the leading analytical combination amplitudes^{6,8} are very accurate even at low temperatures and in clean samples, contrary to claims in Ref. 9.

In conclusion, we have confirmed the validity of our analytical derivation,⁶ which *does* take into account the chemical potential oscillations. We have also shown that the experimentally observed analytical combination amplitudes⁶ of the magnetization are numerically accurate even at zero temperature and even in clean samples. Therefore, the present analysis justifies the use of the analytical theory^{6,8} in the relevant range of all parameters.

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