

New Fundamental dHvA Frequency in Canonical Low-Dimensional Fermi Liquids

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Abstract

We show that a new fundamental period P_f of dHvA oscillations, which appears along with other “forbidden” combination frequencies in a multi-band canonical Fermi-liquid, is very robust with respect to a finite smearing of Landau levels and a background of non-quantized states. We analyse the possibility of measuring small Fermi surface pockets with the use of the “forbidden” frequencies.

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In our previous Letter¹ we predicted qualitatively different dHvA oscillations in a multi-band $2D$ *bulk* metal with a fixed fermion density [canonical ensemble (CE)] compared with an open system where the chemical potential is fixed [grand canonical ensemble (GCE)]. The effect appears because of the pinning of the chemical potential to a fractionally occupied Landau level (LL). Since the chemical potential μ is *constant* across the entire sample, the number of electrons in individual bands would *oscillate* in antiphase if the total number of electrons is kept fixed, as will the partial orbital moments. Therefore, in a closed system there will be a mechanism for different bands to talk to each other producing a dHvA signal with the fundamental frequency, $F = F_1 + F_2 + \dots$, where $F_{1,2,\dots}$ are the ordinary dHvA frequencies of the individual bands.

Let us first briefly recall the qualitative arguments which led us to the prediction of $P_f \equiv 1/F$.¹ In two dimensions the number of electrons which the LL can accommodate ...is always equal to pH , where p is a universal constant [$p = e/(2\pi\hbar c)$]. Then the dHvA resonance condition for fixed n_e is

$$\frac{e(H + \Delta H)(N - 1)}{2\pi\hbar c} = n_e, \quad (1)$$

where n_e is the number of electrons per cm^2 and $N = 1, 2, 3, \dots$ is determined by $eHN/(2\pi\hbar c) = n_e$. Combining these equations, we obtain the fundamental dHvA period

$$P_f \equiv \frac{1}{F} = \frac{p}{n_e}, \quad (2)$$

which is independent of the band structure. This is a period (in $1/H$) with which a fractionally filled Landau level will be completely occupied, independently of any band characteristics.

A relative occupation number of the highest occupied LL is given by $x = F/H - [F/H]$, where $[z]$ stands for the integer part of z . Firstly, it is obvious that this quantity oscillates with frequency F . Secondly, x is relevant for only a *closed* system (Canonical Ensemble) since there the chemical potential is pinned to the highest partially occupied Landau level in the limiting case of negligible damping. Therefore, in the Canonical Ensemble (CE)

the chemical potential oscillates with the same fundamental frequency F as the occupancy x . Since the crystal field is much larger than the magnetic energies involved, oscillations in the chemical potential will cause a charge flow between different sheets of the Fermi surface in a multiband metal. Thus, there will be a component of the orbital magnetic moment oscillating with the fundamental frequency F *interfering* with the standard dHvA individual band frequencies.

This mechanism surely does not operate in a Grand Canonical Ensemble (GCE), where μ is fixed by an external reservoir. Therefore, in the GCE we have just the standard dHvA frequencies. As mentioned in Ref. [1] the pinning of a chemical potential is only possible in 2D. In a 3D metal the Landau levels spread out in bands and the effect disappears together with a pinning of the chemical potential. In general, the effect can be suppressed by any mechanism which introduces a finite density of states (DOS) on the top of a sequence of Landau levels. Therefore the role of the *background* density of states and of the damping should be carefully analyzed.

The background density of states (g_{bkg}) may correspond to either a non-quantized band (e.g. an open orbit) or a heavily damped band, or to the three-dimensional corrections to the energy spectrum, or to a finite contact resistivity. We shall also analyze a truly multiband case (a system with 3 bands with wildly different individual frequencies) which may be relevant for several current experiments. We observe new combination dHvA frequencies appearing because of the chemical potential pinning.

Let us consider a multiband 2D electron gas with a number of bands labeled by the index $\alpha = 1, 2, \dots$. In an external magnetic field H the electronic states will be quantized with energies

$$\epsilon_\alpha(n) = \Delta_\alpha + \hbar\omega_\alpha(n + \frac{1}{2}), \quad n = 0, 1, \dots \quad (3)$$

where $\omega_\alpha = eB/(m_\alpha c)$ is the cyclotron frequency, and $B = H + 4\pi M$ is the internal field. In the following we shall neglect the difference between B and H which is very small because of the small moment in real systems.⁵ Each level is degenerate, contains pH states, and is

broadened by collisions with impurities into a Lorentzian with Dingle width $\sim \hbar/\tau$.^{2,3}

In the Canonical Ensemble ($n_e = \text{const}$) the orbital moment is found from the Helmholtz free energy $\mathcal{F} = \mathcal{F}(n_e, V, T)$ as

$$M_{CE} = - \left(\frac{\partial \mathcal{F}}{\partial H} \right)_{T,V} = -k_B T \int_{-\infty}^{\infty} d\epsilon \left(\frac{\partial g(\epsilon, H)}{\partial H} \right)_{T,V,\mu} \ln \left(1 + e^{(\mu-\epsilon)/k_B T} \right), \quad (4)$$

where $g(\epsilon, H) = g_L(\epsilon, H) + g_{bkg}(\epsilon)$, g_L is the DOS derived from the Landau levels with account for collision broadening,⁴ and $g_{bkg}(\epsilon)$ is the background density of states which may arise because of the above-mentioned reasons. We note that since the Gibbs free energy \mathcal{F} is related to the potential Ω as $\mathcal{F} = \Omega + \mu N$, there is no explicit derivative $\partial\mu/\partial H$ in (4). In the Grand Canonical Ensemble we will find the moment by the use of $\Omega(\mu, T, V)$ as $M_{GCE} = -(\partial\Omega/\partial H)_{T,V,\mu}$. The chemical potential μ is defined by the conservation of the total number of carriers,

$$\int_{-\infty}^{\infty} d\epsilon (g_L(\epsilon) + g_{bkg}(\epsilon)) = n_e \quad (5)$$

To characterize the relative importance of different states, we shall introduce an average density of states, $\bar{g}_L^\alpha = pH/(\hbar\omega_h) = m^\alpha/(2\pi\hbar^2)$, which has the dimension of $eV^{-1}cm^{-2}$. We will then measure the background DOS with respect to \bar{g}_L . Qualitatively, the behavior of μ is quite clear. With the increasing ratio g_{bkg}/\bar{g}_L the oscillatory component of the chemical potential vanishes as will the fundamental frequency. It is important now to quantify the effect of the background DOS, which sets limits on experimental observation. It is also important to analyze a case with a few bands with largely different periods.

From now on we will measure all energies in the natural units of $\beta_0 F \equiv 2\pi\hbar^2 n_e/m$, the moment per area in units of $\beta_0 n_e$, and the thermodynamic potentials in units of $\beta_0 F n_e$, where $\beta_0 = e\hbar/(mc)$.⁶ We shall mark below all dimensionless quantities with a tilde: $\tilde{\mu} = \mu/(\beta_0 F)$, $\tilde{g}_{bkg} = g_{bkg}(\beta_0 F/n_e)$, $\tilde{g}_L^\alpha = m_\alpha/m$.

Let us first consider the case of zero temperature in a clean limit and a constant background DOS. In this case $g_L(\epsilon) = pH \sum_\alpha \sum_{n=0}^{\infty} \delta(\epsilon - \epsilon_\alpha(n))$. Then in the multiband case with the background DOS we will have the following equation for μ in the CE:

$$\sum_{\alpha} N_{\alpha} + x + \tilde{g}_{bkg} \tilde{\mu} = \frac{F}{H}, \quad (6)$$

where N_{α} is the number of completely occupied LLs in a given band. We can write down the following expressions for the thermodynamic potentials, up to an insignificant constant,

$$\begin{aligned} \frac{\mathcal{F}}{\beta_0 F n_e} &= \frac{H}{F} \left(\sum_{\alpha, n}^{occ} \tilde{\epsilon}_{\alpha}(n) + x \tilde{\epsilon}_{\gamma}(N_{\gamma}) \right) + \frac{1}{2} \tilde{g}_{bkg} \tilde{\mu}^2, \\ \frac{\Omega}{\beta_0 F n_e} &= \frac{H}{F} \left(\sum_{\alpha, n}^{occ} (\tilde{\epsilon}_{\alpha}(n) - \tilde{\mu}) + x (\tilde{\epsilon}_{\gamma}(N_{\gamma}) - \tilde{\mu}) \right) - \frac{1}{2} \tilde{g}_{bkg} \tilde{\mu}^2, \end{aligned} \quad (7)$$

where the sum runs over occupied LLs and γ stands for the quantum number of a fractionally occupied LL. We are working in the semiclassical limit, where $N_{\alpha} \gg 1$. In the CE the chemical potential will oscillate about its mean value

$$\tilde{\mu}_{\text{mean}} = \left(1 + \sum_{\alpha} \frac{m_{\alpha}}{m} \tilde{\Delta}_{\alpha} \right) \left(\frac{H}{F} \tilde{g}_{bkg} + \sum_{\alpha} \frac{m_{\alpha}}{m} \right)^{-1}, \quad (8)$$

where m is the bare mass of the electron. The oscillation amplitude of the electron density in individual bands will be $\Delta n_{\alpha}/n_{\alpha} = H/F_{\alpha}$ without background. A background would tend to depin the chemical potential. For depinning of the chemical potential to occur, the background density of states should actually become large, $g_{bkg} \gg g_L$, so that the oscillations of the chemical potential will be reduced as $\Delta\mu \propto H/(F\tilde{g}_{bkg})$. The following calculation substantiates this result.

We compare in Fig. 1 the results of a numerical analysis of a two-band model with mass ratio 1:4 and a band offset $\frac{1}{8}\beta_0 F$. In the top panel we present results for a system without background. The original dependence of a magnetic moment M on the field H is given in the inset, and it looks like a superposition of two sawtooth curves typical of the $M = M(F/H)$ dependence in a one-band case at $n_e = \text{const}$. The fundamental period $F = H + L$ is clearly seen in the Fourier transform, as well as its second and third harmonics ($2F$ and $3F$).⁷ Here H and L stands for the individual dHvA frequencies of a heavy and a light bands, respectively. We also call attention to the presence of other combinational harmonics, though less pronounced, like $H - L$, $F + L$, etc.

With a non-zero background density of states $g_{bkg}/g_L = 5$, the Fourier pattern becomes even better resolved with respect to the presence of a strong fundamental harmonic F ,

together with combination frequencies $H - L$, $F + L$, and $2F - L$ harmonics. We see that although the background is already very large $g_{bkg}/g_L \gg 1$ compared to the light band, it is not strong enough to suppress the fundamental frequency. Only when $g_{bkg}/g_L \sim 25$ do we see that the intensity of a fundamental harmonic reduces below the intensities of individual principal harmonics L and H . Then, with further increase of the background DOS, the combinational frequencies die away and we recover a standard dHvA behavior with two sets of individual harmonics (Fig. 1, bottom panel). It demonstrates that the new fundamental frequency is very robust with respect to the large background density of states.

Another mechanism of reducing the fundamental harmonic would be collision broadening. The effect is observable when $\hbar/\tau \ll (\hbar\omega)_{min}$, which is a standard limitation for the dHvA effect. Therefore, the new effect seems to be very robust and does not require special conditions to be observed.

Now we are going to discuss a 3-band case relevant for current experiments.⁵ This is a two-dimensional Fermi gas in a quantum well with three parabolic bands, where magnetic breakdown is completely unlikely (trajectories are far from the Brillouin zone boundaries) and the Shoenberg effect (magnetic interaction) is negligible.⁵ In this example (Fig. 2) we have considered three bands with the ratio of individual frequencies $F_A : F_B : F_C = 5 : 13 : 53$ and equal masses. Again the fundamental harmonic $F = F_A + F_B + F_C$, marked $F = A + B + C$ in Fig. 2 (top panel), is the second largest after F_C . There is also a rich spectrum of combination frequencies. One could have expected that the contribution of F_A into the overall spectrum to be always very small compared to the B and C bands, irrespective of the ensemble. On the contrary, the band A , although giving a small principal harmonic F_A in CE, produces a very strong combination frequency marked $C - A$ in Fig. 2 (top panel) as it mixes with the band C having largest weight.

Incidentally, this behavior opens up a potential possibility to detect low-frequency modes analyzing “forbidden” dHvA harmonics like $C - A$ in our example. It should be quite easy to detect such a mode by just looking at the positions of the satellite peaks of the strongest harmonics. By attaching the system to a reservoir (in the GCE) one eliminates

all combination “forbidden” frequencies, as shown in Fig. 2 (bottom panel). Comparing CE and GCE one can see a startling difference in behavior of the 3-band system in two regimes, namely, the presence of very extensive combinational harmonics in the Canonical Ensemble (closed system) with the strong involvement of the low-frequency harmonic, which nominally has a very small weight (Fig. 2, top panel).

Referring to other experiments we would like to point out that there are a number of 2D systems, like ET salts, where “forbidden” frequencies may be observed.⁸ Thus, κ -(BEDT-TTF)₂Cu(NCS)₂ salt shows two orbits: α , corresponding to a hole pocket, and β , corresponding to simplest breakdown orbit. “Forbidden” frequencies $F_\beta \pm F_\alpha$ appear on top of the standard ones because of chemical potential pinning, as has been shown within our model by M. Nakano.⁹

Considering Sr₂RuO₄, which we discussed in our previous paper,¹ we have to emphasize the possible relevance of our effect to this interesting 2D metal having the same crystal structure as the high- T_c cuprates. The band structure of Sr₂RuO₄ remains an open issue. While the dHvA model¹⁰ and standard band-structure calculations predict *two* electron and *one* hole pocket,¹¹ recent photoemission spectroscopy results suggest two *hole* pockets¹² as in our example.¹ This difference as well as a significant mass enhancement is presumably due to a strong polaronic shift and the band narrowing as we have discussed for cuprates.^{13,14}

Finally, it is interesting to mention a possible effect of the finite contact resistance R . Contacts are usually made to a sample to measure Shubnikov-de Haas magnetoresistivity. Such a system is apparently “open” and should correspond to a Grand Canonical Ensemble. However, the sample is always capacitively coupled to a contact with some capacitance C . If the ac frequency at which a measurement is carried out is higher than $(RC)^{-1}$ then the exchange by electrons with a reservoir will be suppressed. At these conditions the system will behave in some aspects as being electrically insulated and combination “forbidden” frequencies could be seen.

In conclusion, we have shown that a new dHvA frequency, appearing in the Canonical Ensemble of a two-dimensional electron gas,¹ is very robust with respect to the existence of

smearing factors, like background density of states. The background DOS does not impose any stricter constraints on an observation of the effect compared to the standard (GCE) case. We have demonstrated that the effect produces a tool for identifying small sheets of Fermi surface by analyzing “forbidden” dHvA frequencies. We are also pleased to see that our prediction of the new fundamental dHvA frequency in the canonical multiband 2D Fermi liquid has been confirmed in subsequent Fourier analyses^{7,9} and the relevant experiments are currently underway.⁵

We highly appreciate discussions with D. Shoenberg on our Fourier analysis, with A. Mackenzie on Sr_2RuO_4 , and we thank M.Nakano for sending us his Fourier analysis.⁹ We are grateful to M.Elliott⁵ for information on his recent experiments where combination frequencies have been detected. AMB acknowledges an interesting discussion of our previous Letter¹ with J. Singleton and his group in Oxford, who has found afterwards with co-workers¹⁵ that a “forbidden” frequency $F_\beta - F_\alpha$ in $\kappa\text{-(ET)}_2\text{Cu(NCS)}_2$ salt can be explained by the mechanism that we have suggested earlier.¹ The same conclusion has also been independently reached by M. Nakano.⁹

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FIGURES

FIG. 1. dHvA oscillations in the canonical ensemble with two undamped bands and background density of states g_{bkg} , which is measured w.r.t. the averaged DOS g_L of quantized Landau orbits (see text). A strong signal comes from the fundamental frequency $F = L + H$ when $g_{bkg}/g_L = 0$ (top panel). Relative intensity of the F -harmonic reduces with the increase of a background density of states ($g_{bkg}/g_L = 5$, middle panel), but still remains very strong together with its higher harmonics. These results are to be compared with the Grand Canonical Ensemble (bottom panel) where only standard harmonics are present. Units: $m_H/m_L = 4$, $\Delta_H - \Delta_L = \frac{1}{8}\beta_0 F$, g_L refers to “light” band. Original orbital magnetization is shown in insets.

FIG. 2. dHvA oscillations in the Canonical Ensemble with three undamped bands with frequencies $F_A : F_B : F_C = 5 : 13 : 53$ and equal masses. A large number of combinational frequencies is seen in the Canonical Ensemble (top panel). We note a strong mixing amplitudes $C \pm A$, much stronger than the principal low-frequency harmonic A . The fundamental harmonic obeys a sum rule, $F = F_A + F_B + F_C$ and has the second strongest magnitude with very pronounced higher harmonics ($2F$) and ($3F$). In the Grand Canonical Ensemble the spectrum consists of standard dHvA individual harmonics (bottom panel). Original orbital magnetization is shown in insets.



