

## de Haas–van Alphen Effect in Canonical and Grand Canonical Multiband Fermi Liquid

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A qualitatively different character of de Haas–van Alphen oscillations has been found in a multiband (quasi)two-dimensional Fermi liquid with a fixed fermion density  $n_e$  (canonical ensemble) compared with an open system where the chemical potential  $\mu$  is kept fixed (grand canonical ensemble). A new fundamental period  $P_f$  appears when  $n_e$  is fixed, a damping of the Landau levels is relatively small and a background density of states is negligible.  $P_f$  is determined by the *total* density rather than by the partial densities of carriers in different bands:  $P_f = 1/2n_e\phi$  for spin-split Landau levels and  $P_f = 1/n_e\phi$  in the case of spin degenerate levels where  $\phi$  is the flux quantum.

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The de Haas–van Alphen (dHvA) oscillations of susceptibility as a function of the inverse magnetic field  $1/H$  are well studied both experimentally and theoretically [1]. The frequency of the oscillations is proportional to the extremal cross section of the Fermi surface. Therefore, in the multiband metals one would expect several different dHvA periods corresponding to the independent contributions of different bands [1–3]. The dHvA effect in closed and open systems was analyzed by Dingle back in 1951 [4] with the conclusion that there is practically no difference because the dependence of the chemical potential on the magnetic field, in the case of fixed  $n_e$ , is very weak.

In this Letter we show that while this is true for three dimensions and (or) for the relatively large damping of Landau levels, the dHvA effect is qualitatively different in the near two-dimensional (2D) multiband canonical Fermi liquid compared with the grand canonical ensemble if the damping is relatively weak [5].

If we keep the total number of electrons per area,  $n_e$ , in the near 2D metal fixed, the chemical potential will oscillate with inverse magnetic field,  $1/H$ . These oscillations are responsible for a new fundamental period in the two-band or multiband Fermi liquid. The value of the period can be evaluated by using a simple dHvA resonance condition. There are no partially occupied Landau levels at the resonance. In two dimensions each of the Landau levels can be occupied by  $pH$  carriers per  $\text{cm}^2$ , where  $p = 1/2\phi$  if the levels are spin split, and  $p = 1/\phi$  if they are not ( $\phi = \pi\hbar c/e$  is the flux quantum). 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  $N = 1, 2, 3, \dots$  is determined by

$$\frac{eHN}{2\pi\hbar c} = n_e. \quad (2)$$

Combining Eq. (1) and Eq. (2) we obtain the fundamental dHvA period

$$P_f = \frac{p}{n_e}, \quad (3)$$

which is independent of the band structure according to the following simple argument. In a *multiband* metal the Landau levels (LLs) will be occupied sequentially depending on their energy. The number of electrons which the LL can accommodate is always equal to  $pH$ , where  $p (= e/2\pi\hbar c)$  is constant, i.e., it does not depend on the characteristics of the band. It means that the population of the LLs in multiband metals with field in the canonical ensemble is equivalent to that of a one-band metal. The individual bands will then, generally, show up in an intensity of peaks in susceptibility resulting in additional oscillations of the moment and susceptibility superimposed on the fundamental one. However, the main Fourier component will be the fundamental one with frequency  $1/P_f$ . On the other hand, in the open system this Fourier component is absent or significantly suppressed compared with the individual band oscillations. The conditions for the observation of the fundamental frequency seem to be only marginally stricter, due to its relatively higher value, than those for the ordinary dHvA effect, and it should be observable in near 2D electrically insulated specimens.

It turns out that it is quite difficult to obtain the fundamental frequency in multiband metals with the use of the standard Poisson summation formula. In what follows, we shall therefore consider first the case of a clean 2D metal at zero temperature with (i) the total number of electrons being fixed and (ii) the chemical potential  $\mu$  being fixed (i.e., the metal is well connected to some “reservoir” of electrons). Then we shall estimate the effect of the broadening of the Landau levels.

Let us consider a two-band 2D metal with different band masses, where the bands are split into series of Landau levels (Fig. 1),

$$\epsilon_i(n) = \Delta_i + \hbar\omega_i(n + \frac{1}{2}), \quad n = 0, 1, \dots, \quad (4)$$

where  $i = 1, 2$  is the band index, and  $\omega_i = eH/m_i c$  is the cyclotron frequency. Each level is degenerate, contains  $pH$  states, and is broadened by collisions with impurities into a Lorentzian with Dingle width  $\sim \hbar/\tau$  [4]. We shall assume that  $\hbar/\tau \ll \hbar\omega$ , and describe

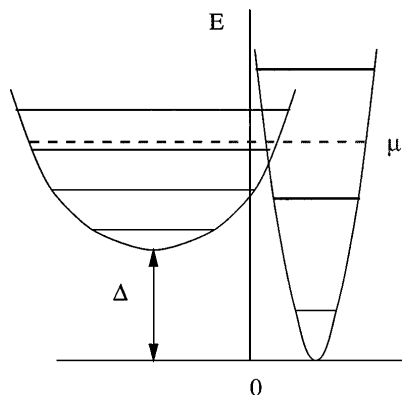


FIG. 1. A schematic representation of the electronic structure of a multiband near 2D metal. The two bands corresponding to heavy and light carriers with the offset  $\Delta$  are shown. In an external magnetic field the bands split into a series of Landau levels whose population depends on the position of the chemical potential  $\mu$ .

the situation where the  $N_1$  levels in the first band and  $N_2$  levels in the second band are occupied, and the last Landau level in the first band is partially occupied by  $xpH$  electrons, where  $x = n_e/pH - [n_e/pH]$ ,  $0 < x < 1$ , and  $[a]$  stands for the integer part of  $a$ .

Let us now consider canonical and grand canonical ensembles.

(i) *Canonical ensemble* ( $n_e = \text{const}$ ). Generally, the orbital moment is found from

$$\begin{aligned} M &= -\left(\frac{\partial F}{\partial H}\right)_{T,V} \\ &= -\frac{2k_B T}{\pi} \int_{-\infty}^{\infty} d\epsilon \text{Im Tr} \frac{\partial G(\epsilon, H)}{\partial H} \ln(1 + e^{(\mu - \epsilon)/T}), \end{aligned} \quad (5)$$

where  $G$  is the electron Green's function which accounts for collisions [6], and the chemical potential  $\mu$  is defined by the conservation of the total number of electrons,  $n_e$  [7]. In the multiband case for a clean metal it would amount to a rather complicated nonlinear equation if we were to apply the standard Poisson summation formula. To elucidate the physics, we shall consider first the limiting case of zero temperature in the clean limit. By counting the number of electrons in the Landau levels we obtain

$$(N_1 - 1)pH + xpH + N_2 pH = n_e, \quad (6)$$

with a similar relation when the LL in the second band is partially occupied. Then we immediately have for the period of the dHvA oscillations  $P_f = 1/H - 1/(H + \Delta H) = p/n_e$ , i.e., the same fundamental period as we have found before, Eq. (3), which is *the same for all bands*. For the energy we have, if the partially occupied LL belongs to the first band,

$$E = \sum_{n_1=0}^{N_1-1} pH\epsilon_1(n_1) + xpH\epsilon_1(N_1) + \sum_{n_2=0}^{N_2} pH\epsilon_2(n_2), \quad (7)$$

with a similar equation when the partially occupied LL belongs to the second band; the moment is found from  $M = -dE/dH$ . We are interested in the semiclassical regime, where the total number of occupied Landau levels is large,  $N_{\text{tot}} = n_e/pH \gg 1$ , as well as the number of occupied LLs in each band,

$$N_i = \left[ \frac{m_i c}{eH} (\mu - \Delta_i) - \frac{1}{2} \right] \gg 1. \quad (8)$$

The chemical potential is pinned to the partially occupied LL and oscillates about the *mean* value  $\bar{\mu} = (2\pi\hbar^2 n_e + \sum_i m_i \Delta_i) / \sum_i m_i$ , which is field independent.

(ii) *Grand canonical ensemble*,  $\mu = \text{const}$ . In that case the period is defined by the condition of the LL crossing the Fermi level,  $\mu = \Delta_i + (e\hbar H/m_i c)(N_i + \frac{1}{2})$ , and we find that the oscillations have independent periods

$$P_i = \frac{e}{m_i c} \frac{1}{\mu - \Delta_i}, \quad (9)$$

with the ratio

$$\frac{P_i}{P_j} = \frac{m_j(\mu - \Delta_j)}{m_i(\mu - \Delta_i)} = \frac{S_i^{-1}}{S_j^{-1}}, \quad (10)$$

where  $S_i$  is the area of Landau orbit in a plane perpendicular to the field. We see that if there were no band offset, the ratio of the periods would have been given by the ratio of inverse masses. If the Dingle temperature is much smaller than the interlevel spacing, the total energy will be given by Eq. (7) without the term containing  $x$ . It is interesting to note the existence of a simple *sum rule* relating individual and fundamental dHvA frequencies

$$\sum_i \frac{1}{P_i} = \frac{1}{P_f}. \quad (11)$$

The moments calculated for two cases with  $n_e = 0.4 \times 10^{14} \text{ cm}^{-2}$ ,  $m_1:m_2 = 1:4$ , and  $\Delta_2 - \Delta_1 = 13 \text{ meV}$  are presented in Fig. 2. It is seen that there is a vast difference between the two regimes. In grand canonical ensemble ( $\mu = \text{const}$ ) the total moment is a sum of two periodic contributions coming with different periods, whereas in the canonical ensemble the moments in the bands follow the fundamental period and change in antiphase following the progressive occupation of the LLs with decreasing field. Although the fundamental period mirrors the level occupation  $x$ , the resulting total moment shows a very complicated behavior which reflects the individual periods with the ratio given by Eq. (10). The reason for this irregularity of the field dependence of the moment (kinks occurring when the current LL is only partially filled) is the *crossing* of the Landau levels belonging to different bands which can only occur when simultaneously  $\Delta_i \neq \Delta_j$  and  $m_i \neq m_j$ . This is because the energy levels in the light band move faster with the field and occasionally the partially occupied LL in the light band sinks below the highest occupied LL in the heavy band and leaves it only partially occupied.

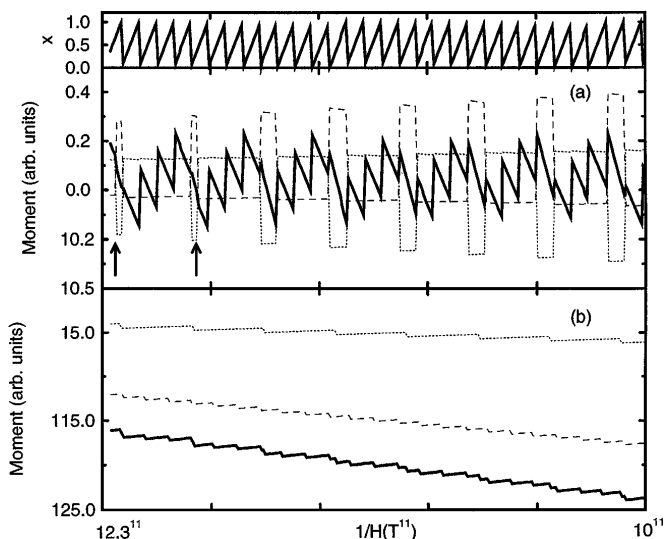


FIG. 2. (a) The dHvA oscillations in the canonical ensemble with two undamped bands,  $m_1:m_2 = 1:4$  and  $\Delta_2 - \Delta_1 = 13$  meV, at the surface density  $n_e = 0.4 \times 10^{14} \text{ cm}^{-2}$ . Arrows indicate the points of Landau level crossing (see text). Bold line: total moment, dotted and dashed lines: partial band contributions. Partial contribution is scaled down to fit into the figure. (b) The dHvA oscillations in the grand canonical ensemble with the same parameters. Bold line: total moment, dotted line: light band, and dashed line: heavy band. In the top panel the filling fraction  $x$  of the partially occupied Landau levels is shown.

This results in discontinuous changes in the moment and susceptibility. The total moment in canonical ensemble oscillates about zero [Fig. 2(a)], similarly to a one-band 2D metal [1]. In the grand canonical ensemble there is a steady flow of electrons into the system with reducing field resulting in an overall monotonous change of the absolute value of the moment [Fig. 2(b)].

The Fourier transform of the total moment (Fig. 3) in the canonical ensemble shows the rich structure of the Fourier components. The light ( $L$ ) band shows up at low frequency with an intensive second harmonic ( $L2$ ) and has a weight much larger compared to the heavy ( $H$ ) band which gives a signal at higher frequency. The fundamental period ( $F$ ) has the largest weight and intensive second ( $F2$ ) and third ( $F3$ ) harmonics. It is easily seen that our *sum rule* (11) holds:  $L + H = F$  ( $= 1.65kT$  in our example).

It is important to consider a situation when the LLs in one band (second, for certainty) are strongly smeared out. Then if the Dingle temperature [4] for the second band is bigger than the LL separation in the first band,  $\hbar/\tau > \hbar\omega_1$ , the second band can be viewed as a uniform background density of states,  $\nu_2(E) \approx \text{const}$ , for the series of the LL in the first band,  $\nu_1(E) = pH \sum_n \delta(E - E_n)$ , where  $\nu_i(E)$  stands for the density of states. One can easily write down the equation for  $\mu$  in this case and apply the Poisson formula to sum over the occupied LLs. This equation can then be solved by successive iterations

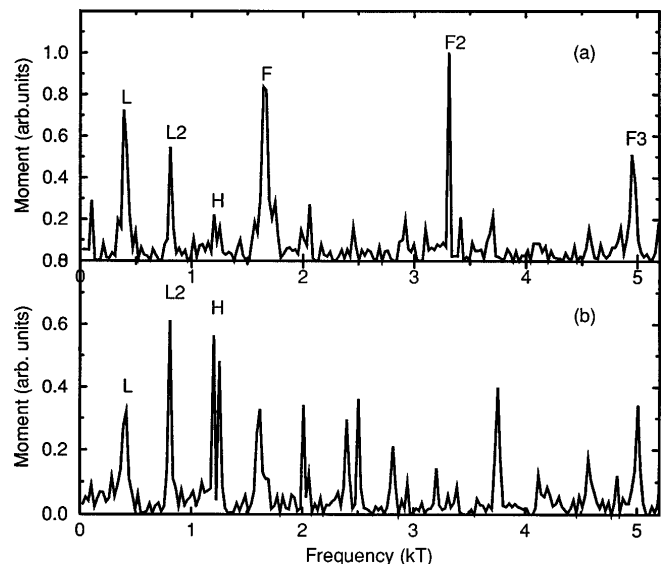


FIG. 3. The Fourier transform of the moment in (a) the canonical ensemble and (b) the grand canonical ensemble. In panel (a) note the clearly resolved components of the light band ( $L$  and the second harmonic  $L2$ ), the heavy band ( $H$ ), and the fundamental period and its higher harmonics ( $F$ ,  $F2$ , and  $F3$ ). The sum rule for the dHvA frequencies holds:  $L + H = F$ . Fundamental frequency equals  $F = 1/P_f = 1.65kT$ . Panel (b) shows standard individual harmonics ( $L$  and  $H$ ) with much less weight in the combined harmonic.

with the result that pinning of the chemical potential at the partially occupied LL vanishes and only the *standard* one-band period,  $P = p/n_1$ , remains. The reason for this is that the second band would work as a reservoir of electrons for the first band and canonical and grand canonical ensembles would become indistinguishable. The same is true if there is a significant background in the density of states. It sets standard constraints for the observation of the fundamental period:  $\max(k_B T, \hbar/\tau_i) < \min(\hbar\omega_i)$ , which could be met in clean samples at low temperatures. We have also performed the calculation for a more realistic model of Landau levels with finite width  $\Gamma_i$  and obtained similar results. The fundamental period is seen in systems obeying this condition and disappears with increasing width  $\Gamma_i$ . In three dimensions each LL develops into a band in such a way that the density of states has a smooth background weakly depending on energy ( $\propto \sqrt{E}$ ). Consequently, the difference between the two ensembles disappears with increasing dimensionality. We expect, therefore, a new fundamental period  $P_f$  to appear when  $n_e$  is fixed; the damping of the Landau levels is relatively small and the background density of states is negligible.

It is worth noting that the magnetic interaction of electrons in different bands could lead to the appearance of a set of combined harmonics like  $mf_1 + nf_2$ , where  $f_i = 1/P_i$  is the dHvA frequency associated with the  $i$ th band and  $m$  and  $n$  are integers (Shoenberg effect) [8,9]. Formally these harmonics would contain the fundamental frequency as well. It is, however, easy to distinguish between the

Shoenberg effect and the effect described here in experiments on insulated and noninsulated samples. In addition, if the magnetic interaction is important, the low frequency difference,  $|f_1 - f_2|$ , may appear as a real oscillation in addition to higher frequencies. One may then expect a characteristic amplitude-modulated signal [9].

The difference between canonical and grand canonical ensembles could be relevant for results of recent dHvA measurements on near 2D  $\text{Sr}_2\text{RuO}_4$  [10]. Three dHvA frequencies of  $3.05kT$ ,  $12.7kT$ , and  $18.5kT$  with the ratio being surprisingly close to 1:4:6 have been found [11]. If they are the genuine frequencies corresponding to three bands as given by band calculations, the fundamental frequency is expected to be  $34.25kT$ . If, on the other hand, we consider only the first two frequencies as being genuine, the fundamental one would be at  $15.75kT$ , which is somewhat smaller than the third measured frequency of  $18.5kT$ . It would be interesting therefore to go into the region of about  $35kT$  to look for the fundamental dHvA frequency in  $\text{Sr}_2\text{RuO}_4$ , and also to compare the dHvA effect in electrically insulated and noninsulated samples.

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