The de Haas - van Alphen effect in two-dimensional metals.

P.D.Grigoriev\textsuperscript{1,2}, I.D.Vagner\textsuperscript{1,3}

\textsuperscript{1}Grenoble High Magnetic Field Laboratory
MPI-FKF and CNRS
BP 166, F-38042 Grenoble Cedex 09, France

\textsuperscript{2}L.D.Landau Institute for Theoretical Physics
142432, Chernogolovka, Moscow region, Russia
e-mail: pasha@itp.ac.ru

\textsuperscript{3}Physics and Engineering Institute at Ruppin ,
Emek Hefer 40250, Israel

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The de Haas - van Alphen effect in two-dimensional (2D) metals is investigated at different conditions and with different shapes of Landau levels (LLs). The analytical calculations can be done when many LLs are occupied. We consider the cases of fixed particle number ($N = const$), fixed chemical potential ($\mu = const$) and the intermediate situation of finite electron reservoir. The last case takes place in organic metals due to quasi-one-dimensional sheets of Fermi surface.

We obtained the envelopes of magnetization oscillations in all these cases in the limit of low temperature and Dingle temperature, where the oscillations can not be approximated by few first terms in the harmonic expansion. The results are compared and shown to be substantially different for different shapes of LLs.

The simple relation between the shape of LLs and the wave form of magnetization oscillations is found. It allows to obtain the density of states distribution at arbitrary magnetic field and spin-splitting using the measurement of the magnetization curve.

The analytical formula for the magnetization at $\mu = const$ and the Lorentzian shape of LLs at arbitrary temperature, Dingle temperature and spin splitting is obtained and used to examine the possibility of the diamagnetic phase transition in 2D metals.

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I. INTRODUCTION

The quantum oscillations of magnetization and conductivity in magnetic field (the de Haas - van Alphen and Shubnikov - de Haas effects) give abundant information about electronic structure of metals.\textsuperscript{1} The theory of these effects in three-dimensional (3D) metals has been constructed\textsuperscript{2} and proved well by many experiments\textsuperscript{1}. However, this theory is not applicable in two-dimensional (2D) compounds, when warping of the Fermi surface (FS) is less than the Landau level separation $\hbar \omega_c$. The calculation of the kinetic properties of 2D compounds in magnetic field is difficult even in the simplest model approximations\textsuperscript{3,4}. Including of the electron-electron interaction, other parts of FS or different scattering mechanisms makes the problem formidable and the quantitative theory of the 2D Shubnikov - de Haas effect does not exist. The theory of the de Haas - van Alphen (dHvA) effect is much simpler because the magnetization is a thermodynamic quantity. The complete description of the 2D dHvA effect, however, does not exist either. There are two approaches to this problem. The first is to make the harmonic expansion of magnetization oscillations\textsuperscript{2}, in analogy to the Lifshitz-Kosevich formula\textsuperscript{2}. This expansion assumes the chemical potential to be fixed ($\mu = const$). In most compounds it is the electron density which is constant and the chemical potential is an oscillating function of the magnetic field. Even at fixed chemical potential this approach in pure metals at low temperature gives the infinite harmonic series for magnetization oscillations with slow convergence which is not convenient for the practical use. This series has been summed explicitly only at zero temperature and the Lorentzian shape of LLs\textsuperscript{1,2}.

Another approach is the direct summation over Landau levels (LLs). It is applicable to the systems with fixed electron density ($N = const$) and just at low temperature $T \ll \hbar \omega_c$ ($\hbar \omega_c = \hbar eB/m^*c$ is the LL separation) since then only few LLs near FS are important\textsuperscript{3}. The analytical calculations of the magnetization and its envelope by this method at low but nonzero temperature have been done only for sharp Landau levels (LLs). This approximation has a very narrow domain of applicability $\hbar \omega_c \gg$
We study the case of constant chemical potential because the Landau level broadening $\Gamma$ has a very strong influence on the magnetization oscillations. Any electron reservoirs are also ignored in these calculations although in organic metal the reservoir of field independent electron states exists due to quasi-one-dimensional parts of FS. The influence of this electron reservoir on the magnetization oscillations has been studied recently only for sharp LLs. Numerical calculations are possible [10], [11], but the numerical analysis is not evident and convenient for practical use. So, in many cases one continues to apply the Shoenberg’s 2D harmonic expansion.

Our work is intended to study the 2D dHvA effect with arbitrary shape and width of the LLs and under conditions close to real experiments.

In sections II, III, IV we consider the case of fixed electron density using the method of direct summation over LLs. The reservoir electron states and finite temperature are also taken into account. In Sec. II we derive the equation for the chemical potential and the relation between the magnetization and the chemical potential that are valid at arbitrary shape and width of LLs. In section III this is used to derive the simple relation between magnetization oscillations and the density of states (DoS) function. We describe in details how this relationship enables the direct measurement of the DoS distribution.

The problem of the density of states distribution in 2D metals in magnetic field is an important and old one. Since the discovery of the integer quantum Hall effect many theoretical approaches have been developed to solve it (for example, [3], [4]) and it would be very interesting to compare the results of these calculations with the experimentally measured DoS distribution. The DoS function tells about the electron spectrum and the scattering mechanisms that determine the kinetic phenomena but it is difficult to explicitly relate the DoS distribution to the conductivity. The magnetization gives a quite direct information about the DoS and an accurate method to extract the DoS distribution from the magnetization oscillations is needed. The harmonic expansion is not good here for the following reasons. To obtain the DoS distribution function one should consider the case of constant electron density and low temperature when many harmonics are important. In this case the explicit harmonic expansion (with many terms) of magnetization oscillations does not exist and the influence of LL broadening on the harmonic damping can not be separated accurately from other factors. The method of extracting the DoS distribution from the dHvA measurements has been proposed by M. Ya. Azbel [12]. This method is valid only in the extremely strong magnetic field when the magnetic length $l_0 = (\hbar c/eB)^{1/2}$ is much less than the impurity Bohr radius. His procedure assumes that one can realize the situation when only one LL is occupied (this means that $\hbar \omega_c/2 > E_F$, $E_F$ is the Fermi energy). Such a condition can not be fulfilled in organic metals and many other compounds where Fermi energy is quite great. Azbel’s procedure also assumes zero temperature, no electron reservoir and no warping of Fermi surface. Our approach has no these defaults and limitations.

In Sec. IV we obtain the analytical formulas for the envelopes of the magnetization oscillations. In Sec. V we study the case of constant chemical potential $\mu = const$ using the harmonic expansion. We obtain the analytical formula for the magnetization with Lorentzian shape of LLs for arbitrary temperature, Dingle temperature and spin splitting. This formula is used to examine the possibility of the diamagnetic phase transition in 2D metals. We also derive the relations between the oscillations of the magnetization and the oscillations of the chemical potential (in the case $N = const$) or the particle density (when $\mu = const$) at general conditions and find their domain of applicability.

Both the conditions $N = const$ and $\mu = const$ take place in real compounds that was determined on the experiment using the shape of magnetization oscillations [3], [7], [4] or by the observation of $\beta - \alpha$ frequency [3], [4] in the presence of the magnetic breakdown. Therefore we consider both these cases.

II. DERIVATION OF MAGNETIZATION IN THE CASE $N=\text{const}$ WITH FINITE ELECTRON RESERVOIR

The condition of the constant particle density ($N = \text{const}$) means that the sample is isolated from electron reservoirs and the sample volume is constant. This takes place in heterostructures [3] and is usually more realistic than restriction of the fixed chemical potential ($\mu = \text{const}$). Nevertheless, in many solids the exact conditions $N = \text{const}$ or $\mu = \text{const}$ are not fulfilled. The intermediate situation takes place, for example, in synthetic metals like intercalated graphite compounds where the magnetic field dependent charge transfer between the graphite layers and intercalated layers takes place, and in organic metals due to the additional quasi-1D parts of FS, that does not contribute to the magnetization directly.
but serve as a finite electron reservoir. The influence of this electron reservoir on the magnetization oscillations has been studied recently \[9\] only for sharp LLs and in the main part numerically. We shall consider arbitrary shape of LLs.

Since the total particle number remains constant, this intermediate situation can be considered as the case \(N = \text{const}\) with some additional density of states (DoS). This reservoir part of the DoS does not oscillate with magnetic field and changes on the scale of Fermi energy. Since only several LLs near FS are important for the dHvA oscillations and the number of occupied LLs in organic metals \(n_F \gg 1\) this reservoir part of DoS may be considered as constant. For the same reason we assume the LL degeneracy be equal for all LLs. Then the total density of electron states may be written in the form

\[
\rho(\varepsilon, B) = g(B) \sum_{n=0}^{\infty} D_n(\varepsilon - \hbar \omega_c(n + \frac{1}{2})) ,
\]

where the LL degeneracy \(g(B) = 2B/\Phi_0 + n_R \cdot \hbar \omega_c\) includes two spin polarizations and the reservoir density of states \(n_R = \text{const}\), \(\Phi_0 = 2\pi \hbar c/e\) is the flux quantum. \(D_n(\varepsilon)\) is normalized to unity and gives the shape of the \(n\)-th LL with some spin-splitting and the reservoir part of the DoS.

To calculate the magnetization we need first to calculate the chemical potential. It is given by the equation

\[
N = \int \frac{\rho(\varepsilon, B) d\varepsilon}{1 + \exp \left(\frac{\varepsilon - \mu}{T}\right)}
\]

It is convenient to count the energy and the chemical potential from the last occupied LL:

\[
\delta \mu \equiv \mu - \hbar \omega_c(n_F + 1/2) ; \quad E \equiv \varepsilon - \hbar \omega_c(n_F + 1/2)
\]

where \(n_F = \text{Int}[E_F/\hbar \omega_c]\) is the number of completely filled LLs when \(\delta \mu = 0\). It is an integer and jumps by unity one time on each dHvA period.

Now we apply the direct summation over the LLs. Another approach of the harmonic expansion of the DoS \[8\] will be used in the next section for the case \(\mu = \text{const}\). At \(T \ll \hbar \omega_c\) only three LLs near FS produce the temperature dependent contribution to the oscillations because the contribution from other LLs is small by a factor \(\exp^n(-\hbar \omega_c/T)\). The equation \[3\] then simplifies

\[
\tilde{n} + 1 = \int_{-3\hbar \omega_c/2}^{3\hbar \omega_c/2} \frac{D(E) dE}{1 + \exp \left(\frac{E - \delta \mu}{T}\right)}
\]

where \(\tilde{n} = N/g - n_F = F/B - n_F\) is the filling factor of the last occupied LL, \(N\) is the area electron density, \(F = \text{const}\) is the dHvA frequency. \(\tilde{n}(B)\) is an oscillating function of the magnetic field. On each dHvA period it monotonically decreases from 1 to 0 with increasing of the magnetic field. The DoS function \(D(E)\) here is periodic \(D(E + \hbar \omega_c) = D(E)\) and normalized to unity

\[
\int_{-\hbar \omega_c/2}^{\hbar \omega_c/2} D(E) dE = 1
\]

The LLs just above and just below the last occupied LL are important when the chemical potential is situated just between two LLs \(|\delta \mu| - \hbar \omega_c/2 \sim T\). Then these two LLs are in equal position and hence have the same shape. Therefore, we have written the same DoS function \(D(E)\) for all three LLs.

Now we have to calculate the thermodynamic potential. By definition

\[
\Omega = -k_B T \int d\varepsilon \rho(\varepsilon, B) \ln \left(1 + \exp \left(\frac{\mu - \varepsilon}{k_B T}\right)\right),
\]

where the density of states is given by \[1\]. As in the derivation of \[4\] we calculate the temperature dependent contribution only from three LLs near FS. Actually we could leave only one LL but now the derivation is more general. The expression for the thermodynamic potential now becomes

\[
\Omega = -T g \sum_{n=0}^{n_F-2} \frac{\mu - \hbar \omega_c(n + 1/2)}{T} -
\]
-\theta g \int_{-3\hbar \omega_c/2}^{3\hbar \omega_c/2} dE D(E) \ln \left( 1 + \exp \frac{\delta \mu - E}{T} \right)

The summation in the first term is easy and gives \( g \hbar \omega_c (n_F - 1)^2 / 2 - g \mu (n_F - 1) \). We need the free energy

\[ F = \Omega + \mu N = g \mu (\bar{n} + 1) + g \hbar \omega_c \frac{(n_F - 1)^2}{2} - \theta g \int_{-3\hbar \omega_c/2}^{3\hbar \omega_c/2} dE D(E) \ln \left( 1 + \exp \frac{\delta \mu - E}{T} \right) \]

(7)

To obtain the magnetization \( M = -dF/dB \) one should differentiate all the magnetic field dependent quantities in this expression. As a result one obtains a quite huge formula. We do not write down it here because it is useless for the analytical calculations. Even for numerical calculations it is simpler to use (7). The elegant expression for the magnetization can be obtained if many LLs are occupied (\( n_F \gg 1 \)). This condition is always satisfied in organic metals where \( n_F \sim 300 \). Then in the third term of (7) one can differentiate only oscillating quantities since they give additional factor \( n_F \). The only such a quantity is \( \delta \mu \). We assume the function \( D(E) \) to be independent of the position of the chemical potential, that is usually well satisfied. The terms with \( \frac{d\delta \mu}{dB} \) in \( dF/dB \) cancel each other after using (3) and (4). As a result we get

\[ M \approx C \left[ \frac{1}{2} - \bar{n} + \frac{\delta \mu}{\hbar \omega_c} \right] \]

(8)

where \( \delta \mu \) is the solution of (4). The prefactor

\[ C \equiv \frac{\theta}{B} \hbar \omega_c n_F \approx \frac{\theta}{B} E_F = \text{const} \]

(9)

is the same for different dHvA periods since \( \theta \sim B \). The first two terms in square brackets in (8) give the sawtooth form of magnetization, and the last term \( \frac{\delta \mu}{\hbar \omega_c} \) determines the damping of the oscillations due to finite temperature and the LL broadening. The expression in square brackets of (8) is the oscillating part of the chemical potential divided by \( \hbar \omega_c \). This can be easily shown using the definition (3) of \( \delta \mu \). Then the formula (8) becomes

\[ M = C (\mu - E_F) / \hbar \omega_c \]

(10)

Another (more apparent) derivation of this relation between the oscillations of the magnetization and the chemical potential (formula (56)) and its discussion will be given in section IV.

So, in this section we have derived the formulas (4) for the chemical potential and (8) for magnetization at finite temperature and arbitrary DoS distribution. The density of electron reservoir states was assumed to be approximately constant on the scale of \( \hbar \omega_c \). In the next two sections we shall use these formulas to obtain more concrete results that are useful for the analysis of the dHvA effect.

III. RELATION BETWEEN THE MAGNETIZATION AND THE DOS FUNCTION

Now we derive the relation between the wave form of the the magnetization \( M(B) \) and the DoS distribution \( D(E) \), that allows the direct measurement of the function \( D(E) \) at different magnetic field values and spin-splitting if the temperature smearing is much less than the LL broadening. We assume \( T \ll \Gamma \), where \( \Gamma \) is the width of LLs. Since we need the function \( M(B) \) on one particular dHvA period \( (0 < \bar{n} < 1) \) it is convenient to consider magnetization as a function of \( \bar{n} \approx n_F (B_0 - B)/B_0 \), where on this period \( \bar{n}(B_0) = 0 \). Differentiating (8) we obtain

\[ \frac{dM}{d\bar{n}} = C \left[ -1 + \frac{1}{\hbar \omega_c} \frac{d\delta \mu}{d\bar{n}} \right] \]

(11)

Differentiating the equation (8) with respect to \( \delta \mu \) we get

\[ \frac{dM}{d\bar{n}} = C \left[ -1 + \frac{1}{\hbar \omega_c} \frac{d\delta \mu}{d\bar{n}} \right] \]
\[
\frac{d\tilde{n}}{d\delta \mu} = \int_{-3\hbar \omega_c/2}^{3\hbar \omega_c/2} \frac{D(E) \ dE}{4T \cosh^2 \left( \frac{E-\delta \mu}{2T} \right)} \tag{12}
\]

At \( T \ll \Gamma \) the important region of the integration is \( |E-\delta \mu| \sim T \) and the function \( D(E) \) can be expanded in Taylor series near this point

\[
\frac{d\tilde{n}}{d\delta \mu} = \int_{-3\hbar \omega_c/2}^{3\hbar \omega_c/2} \frac{D(\delta \mu) + D''(\delta \mu) \frac{(E-\delta \mu)^2}{2} + \ldots \ dE}{4T \cosh^2 \left( \frac{E-\delta \mu}{2T} \right)} \tag{13}
\]

The all odd terms drop out because the integrand should be a symmetric function of \((E-\delta \mu)\). We shall keep only the \( T^2 \) term. The integration can be extended to infinity. After the integration we get

\[
\frac{d\tilde{n}}{d\delta \mu} = D(\delta \mu) + T^2 D''(\delta \mu) \frac{n^2}{6} + O(T^4) \tag{14}
\]

Substituting this into (11) we obtain

\[
\frac{dM}{d\tilde{n}} = C \left[ -1 + \frac{1}{\hbar \omega_c \left( D(\delta \mu) + T^2 D''(\delta \mu) \frac{n^2}{6} + O(T^4) \right)} \right] \tag{15}
\]

The temperature dependent terms in (13) are small and can be separated by taking the measurements at several low temperatures and extrapolating to \( T = 0 \). So, one can measure the function

\[
\frac{dM_{T=0}(\tilde{n})}{d\tilde{n}} = C \left[ -1 + \frac{1}{\hbar \omega_c D(\delta \mu)} \right] \tag{16}
\]

On the experiment it is very difficult to obtain the proportionality coefficient between the measured signal and the magnetization. For this reason we shall look at the constant \( C \) as on an unknown factor and later we shall describe how one can obtain it. Now suppose one can measure the ratio

\[
R(\tilde{n}) \equiv \frac{M_{T=0}(\tilde{n})}{C} \approx \left[ \frac{1}{2} - \tilde{n} + \frac{\delta \mu}{\hbar \omega_c} \right] \tag{17}
\]

With this function one can rewrite the equation (16) as

\[
D(\delta \mu(\tilde{n})) = \frac{1}{\hbar \omega_c \left( \frac{dR(\tilde{n})}{d\tilde{n}} + 1 \right)} \tag{18}
\]

The function \( D(\delta \mu(\tilde{n})) \) is not \( D(\delta \mu) = D(E) \). The dependence \( \delta \mu(\tilde{n}) \) is simply related to the function \( R(\tilde{n}) \) since the formula (17) can be casted into

\[
\delta \mu(\tilde{n}) = \hbar \omega_c \left[ \tilde{n} - \frac{1}{2} + R(\tilde{n}) \right] \tag{19}
\]

The DoS distribution \( D(E) \) is just the plot of \( D(\tilde{n}) \) (18) as a function of \( \delta \mu(\tilde{n}) \) (19).

Now we have to say how one can get the constant \( C \) by which the measured torque should be divided to obtain the function \( R(\tilde{n}) \) (17). The normalization condition (3) does not give this constant since for any \( C \) the described procedure gives automatically normalized DoS distribution. The constant \( C \) determines the strength of the magnetization and, hence, the DoS oscillations. The smaller one takes the constant \( C \) in (17), the larger oscillations of the DoS function he obtains. There is some critical value \( C_0 \) such that if one assumes \( C < C_0 \) he gets a singularity in the DoS, calculated from (18). At \( C = C_0 \) this singularity is in the middle of a LL. The peak value \( D(0) \) of the DoS is indeed large compared to the average DoS \( \bar{D} = 1/\hbar \omega_c \) for small LL broadening \( \Gamma \ll \hbar \omega_c \). The correct value of the constant \( C \) is then close to \( C_0 \) which gives the function \( D(E) \) accurately always except the vicinity of the peaks of LLs. We shall use this singularity to obtain more accurate value of the constant \( C \).

The derivative (19) has a sequence of the periodically situated minima that occur on each dHvA period when the chemical potential crosses the position in the middle of a LL \((\delta \mu = 0)\). Since \( n_F \gg 1 \) and the
magnetization is measured on many dHvA periods these minima form a smooth function of magnetic field

\[ M'_{\text{min}}(B) = C \left[ -1 + \frac{1}{\hbar \omega_c D(0)} \right] \]

This function monotonically decreases to some finite limit since \( \hbar \omega_c D(0) \) decreases with the increasing of the magnetic field. This is because \( \hbar \omega_c \sim B \) while the peak value of the DoS \( D(0) \) has the very slow dependence on the magnetic field. In the vicinity of the peaks the DoS distribution may be given by only two parameters: the width of the LL \( \Gamma \) and the peak value \( D(0) = 1/\alpha \Gamma \). The width \( \Gamma \) is determined mainly by the impurity scattering which is approximately independent of the magnetic field. The parameter \( \alpha \sim 1 \) depends on the DoS distribution but also is almost independent of the magnetic field. For Lorentzian shape of LLs \( \alpha = \pi \).

Hence, one can assume \( D(0) = 1/\alpha \Gamma = \text{const} \) and from the curve of the minimum values of

\[ M'_{\text{min}}(B) = C \left[ -1 + \frac{\alpha \Gamma}{\hbar \omega_c} \right] \]  \hspace{1cm} (20)

one can easily obtain two unknown constants: \( \alpha \Gamma \) and \( C \). This value of the constant \( C \) is accurate enough to calculate \( D(E) \) even in the vicinity of the maxima.

The above procedure gives the DoS at the position of the chemical potential while the magnetic field and the chemical potential are varied. In general, this is not the same as if one fix the magnetic field and the chemical potential but vary the energy itself. In our analysis we disregard the dependence of \( D(E) \) on the position of the chemical potential and hence this difference. This approximation works when the DoS distribution is determined by one-electron processes. These are electron scattering on lattice imperfections and inhomogeneities, the finite probability of the interlayer jumping and so on. When the many particle effects play an important role (for example, the change of the magnetic field drives a sequence of phase transitions as in fractional quantum Hall effect), our results are not applicable.

We have neglected the nontrivial many body effects at the beginning when have written the formulas (1) and (6) assuming that the system may be described by the distribution of the single fermion states \([6]\). Nevertheless, this has a very wide application region because usually fine many particle effects are damped by impurities, finite \( k_z \) dispersion and other factors, especially when many LLs are occupied.

One can not get the value of LL separation \( \hbar \omega_c \) without performing some more measurements because after we have neglected the temperature dependent terms, the LL separation becomes the only parameter with the dimensionality of energy. To find it one needs to compare it with some other energy parameter. If one knows the g-factor of the spin-splitting in this compound he can compare \( \hbar \omega_c \) with the spin-splitting energy. One can also use the temperature dependence of the amplitude of magnetization oscillations to obtain \( \hbar \omega_c \), but this would be less accurate than the cyclotron resonance method. So, there are many ways to get \( \hbar \omega_c \).

To summarize, the suggested procedure of extracting of the DoS distribution from the oscillations of magnetization consists of the two steps. The first is to obtain the constant \( C \) (that normalizes the measured signal) and the second to plot \( D(E) \) itself. To do this one should measure the torque as a function of \( x = F/B \) where \( F = \text{const} \) and \( x \) changes by unity on each dHvA period. Then \( x = n_F + \tilde{n} \).

This function \( M(x) \) should be measured at several low temperatures and extrapolated to \( T = 0 \). As a result one gets an oscillating function \( M_0(x) \) with period equal to unity. We also need its derivative \( M'_0(x) = dM_0(x)/dz \) which is also an oscillating function. The minima of \( M'_0(x) \) form a smooth monotonic function \( M_{\text{min}}(x) \). Extrapolating this function to \( x = 0 \) one gets the constant

\[ C = \lim_{x \to 0} M_{\text{min}}(x) \]

Or one can use the formula \([20]\) and obtain the constant \( C \) from \( M_{\text{min}}(x) \) more accurately. Substituting the functions \( R(x) = M_0(x)/C \) and \( R'(x) = M'_0(x)/C \) into \([18]\) and \([19]\) one can plot the expression \([18]\) as a function of \([14]\) on one period of the oscillations. This plot is the desired DoS distribution at the magnetic field corresponding to the chosen dHvA period.

The described procedure allows to measure the DoS distribution on the Fermi level for different magnetic field values (because one can obtain \( D(E) \) on each dHvA period) and for different spin-splitting energy (because one can tilt the magnetic field with respect to the conducting plane of a sample). This
information about the DoS distribution is very important for the study the role of different scattering mechanisms in the electron motion at different external parameters. Such an information can not be obtained using the previous methods of processing the data of the dHvA measurements where one assumed some particular shape $D(E)$ and then numerically calculated the magnetization. The results of the calculation (usually only of the envelope) one compared with the measured signal and if the agreement was good enough the chosen function $D(E)$ was taken as a result. Our method is more simple and accurate.

IV. THE ENVELOPE OF MAGNETIZATION OSCILLATIONS

We can also calculate the envelope of magnetization oscillations for several simple DoS distributions in the limit $\bar{\hbar}\omega_c \gg \Gamma \gg T$. At low temperature the envelope turns out to depend strongly on the shape of the LLs while the additional constant contribution to the DoS from other parts of FS almost does not change the envelope. The envelope of magnetization oscillations does not give as much information about electronic structure of the compounds as the DoS distribution does. Nevertheless it is still useful for the analysis of the dHvA effect and is quite easy for measurement.

To calculate the envelope of magnetization oscillations we shall take the total density of states function in the form

$$D(E) = (1 - \kappa) \frac{D_0(E/\Gamma)}{\Gamma} + \frac{\kappa}{\hbar\omega_c}$$

where $\Gamma$ is the width of LLs, $\kappa < 0$ is a number that determines the constant part of the DoS and

$$\int_{-\hbar\omega_c/2}^{\hbar\omega_c/2} D_0 \left( \frac{E}{\Gamma} \right) \frac{dE}{\Gamma} = 1$$

converges rapidly. Then the function $D(E)$ is also normalized to unity. The degeneracy $g(B)$ of LLs should be renormalized to include all additional parts of FS.

How does the envelope of magnetization depend on $\kappa$. The answer is that in the limit $n_F \gg 1$ the constant part of DoS affects only the shape of magnetization but not the envelope of the oscillations. This is different from the case $\mu = const$ where the constant part of DoS does not change both the envelope and the shape of the oscillations.

To show this we first should substitute (21) into (4). The LLs just above and just below the last occupied one contribute only when $|\delta\mu| \approx \hbar\omega_c/2$. As we shall see, if $T, \Gamma \ll \hbar\omega_c$ the extrema of magnetization take place when $|\delta\mu| \ll \hbar\omega_c/2$. So, the regions where $(\hbar\omega_c/2 - |\delta\mu|) \sim T \ll \hbar\omega_c/2$ are not important for the envelope of magnetization and $T$ and $\Gamma$ dependent contribution is given by only one LL. Then the equation (4) becomes

$$\tilde{n} = \int_{-\hbar\omega_c/2}^{\hbar\omega_c/2} \frac{D(E)}{1 + \exp \left( \frac{E - \delta\mu}{T} \right)}$$

The integral

$$\int_{-\hbar\omega_c/2}^{\hbar\omega_c/2} \frac{dE}{1 + \exp \left( \frac{E - \delta\mu}{T} \right)} \approx \frac{\hbar\omega_c}{2} + \delta\mu$$

and after substitution of (21) the equation (22) acquires the form

$$\tilde{n} = \int_{-\hbar\omega_c/2}^{\hbar\omega_c/2} \frac{(1 - \kappa)D_0(E)}{1 + \exp \left( \frac{E - \delta\mu}{T} \right)} + \kappa \left( \frac{1}{2} + \frac{\delta\mu}{\hbar\omega_c} \right)$$

Substituting this into the expression (8) for the magnetization we get

$$M = \frac{g^*}{B} \hbar\omega_c n_F \left[ \frac{1}{2} + \frac{\delta\mu}{\hbar\omega_c} - \tilde{n} \right]$$
where \( g^* = (1 - \kappa)g \) is the LL degeneracy of only the oscillating part of DoS and

\[
\tilde{n}_0(\delta \mu) = \int_{-\hbar\omega_c/2}^{\hbar\omega_c/2} \frac{D_0(E) dE}{1 + \exp \left( \frac{E - \delta \mu}{T} \right)}
\]  

(25)

The magnetization on each dHvA period may be considered as a function of \( \delta \mu : M(B) = M(\delta \mu) \). The envelope of the magnetization oscillations is then given by

\[
M_{\pm}(B) = M(\delta \mu_{ex}(B)) = C^* \left[ \frac{1}{2} + \frac{\delta \mu_{ex}}{\hbar \omega_c} - \tilde{n}_0(\delta \mu_{ex}) \right]
\]  

(26)

where \( C^* = g^* \hbar \omega_c n_F / B \) and the extremum values \( \delta \mu_{ex}(B) \) at which the magnetization has maxima or minima are given by the equation

\[
\frac{dM}{dB} = 0 \Leftrightarrow \frac{dM(\delta \mu)}{d\delta \mu} = C^* \left( \frac{1}{\hbar \omega_c} - \frac{d\tilde{n}}{d\delta \mu} \right) = 0
\]  

(27)

After accounting for (23) this equation becomes

\[
\frac{d\tilde{n}}{d\delta \mu} = \int_{-\hbar\omega_c/2}^{\hbar\omega_c/2} \frac{(1 - \kappa)D_0(E) dE}{4T \cosh^2 \left( \frac{E - \delta \mu_{ex}}{2T} \right)} + \frac{\kappa}{\hbar \omega_c} = \frac{1}{\hbar \omega_c} \Rightarrow
\]

\[
\int_{-\hbar\omega_c/2}^{\hbar\omega_c/2} \frac{D_0(E) dE}{4T \cosh^2 \left( \frac{E - \delta \mu_{ex}}{2T} \right)} = \frac{1}{\hbar \omega_c}
\]  

(28)

The equation (28) for \( \delta \mu_{ex} \) is independent of \( \kappa \) and so does the envelope of magnetization given by (26).

The function \( \delta \mu(B) \) is monotonic on each dHvA period and different on different dHvA periods. Moreover, if \( D(E) \) is a symmetric function then \( \delta \mu \) is an antisymmetric function of \( (\tilde{n} - 1/2) \) that can be obtained from (4) using the identity

\[
\frac{1}{1 + \exp \frac{E - \delta \mu}{T}} = \frac{1}{2} - \frac{1}{2} \sinh \frac{E}{T} - \sinh \frac{\delta \mu}{T} \cosh \frac{E}{T}
\]

The magnetization (8) then is also an antisymmetric function of \( (\tilde{n} - 1/2) \).

In the limit \( T \ll \Gamma \) the function \( D_0(E) \) may be expanded in Taylor series as has been done in (13). If we rest only \( \sim T^2 \) terms the equation (28) becomes

\[
D_0(\delta \mu_{ex}) + T^2 D_0'(\delta \mu_{ex}) \frac{\pi^2}{6} = \frac{1}{\hbar \omega_c}
\]  

(29)

Formula (25) can also be simplified. Integrating by parts and expanding up to the terms \( \sim T^2 \) we get

\[
\tilde{n}_0 = G_0(\delta \mu) + T^2 D_0'(\delta \mu) \frac{\pi^2}{6}
\]  

(30)

where we have introduced the function

\[
G_0(E) = \int_{-\hbar\omega_c/2}^{E} D_0(E') dE'
\]  

(31)

The function \( G_0(E) \) is dimensionless and changes in the range \([0; 1]\). From the equation (28) one can immediately say that if \( D_0(x) \) has the exponentially falling tails, the \( \delta \mu_{ex} \sim \pm \Gamma \ln(\hbar \omega_c / \Gamma) \). If \( D_0(x) \) falls off as \( x^{-\gamma} \), then \( \delta \mu_{ex} \sim \Gamma(\hbar \omega_c / \Gamma)^{1/\gamma} \). This determines the behavior of the envelope (26). Now we shall consider in details two different functions \( D_0(x) \).
A. Exponentially falling $D(x)$

There are many symmetric exponentially falling functions that are eligible to be a density of states function $D_0(E)$. They all lead to the close expressions for the envelope of the magnetization oscillations and for example we shall take

$$D_0(E) = \frac{1}{4\Gamma \cosh^2 \left( \frac{E}{2\Gamma} \right)} \quad (32)$$

Now the equation (28) possesses the symmetry $T \leftrightarrow \Gamma$ if $T \ll \hbar \omega_c$ and one can easily obtain the limit $\Gamma \ll T$ from the limit $T \ll \Gamma$.

The equation (29) can be solved by the iteration procedure with small parameter $T^2/\Gamma^2$. In zeroth approximation

$$D_0(\delta \mu_{ex}^0) = \frac{1}{4\Gamma \cosh^2 \left( \frac{\delta \mu_{ex}^0}{2\Gamma} \right)} = \frac{1}{\hbar \omega_c} \quad (33)$$

Since the magnetization (8) is an antisymmetric function of $\delta \mu$ we shall consider only one (negative) root of this equation

$$\delta \mu_{ex}^0 = -2\Gamma \arccosh \sqrt{\frac{\hbar \omega_c}{4\Gamma}} \approx -\Gamma \ln \left( \frac{\hbar \omega_c}{\Gamma} \right) \quad (34)$$

From equation (30) we get

$$\tilde{n}_0(\delta \mu_{ex}^0) \approx \frac{1}{1 + \frac{\hbar \omega_c}{\Gamma}} \quad (35)$$

Substituting (33) and (34) into (29) we get the envelope in zeroth approximation

$$M_0^\pm = \pm C^* \left[ 1 - \frac{\Gamma}{\hbar \omega_c} \ln \left( \frac{\hbar \omega_c}{\Gamma} \right) - \frac{\Gamma}{\hbar \omega_c} \right] \quad (35)$$

If one makes the replacement $\Gamma \rightarrow T$ this result coincides with the result of Vagner et al. [7], obtained for the case $\Gamma = 0$ and finite temperature. It’s not surprising because of the mentioned symmetry $\Gamma \leftrightarrow T$ with the DoS function (32).

One can easily obtain the first temperature correction to the envelope (35). In the first approximation the equation (29) becomes

$$D_0(\delta \mu_{ex}^0) + T^2 D_0''(\delta \mu_{ex}^0) \frac{\pi^2}{6} = \frac{1}{\hbar \omega_c} \quad (36)$$

The correction to the chemical potential

$$\Delta T \mu_{ex} \equiv \delta \mu_{ex}^1 - \delta \mu_{ex}^0 = \frac{T^2 D_0''(\delta \mu_{ex}^0) \pi^2}{6 D_0'(\delta \mu_{ex}^0)} \quad (37)$$

This correction $\Delta T \mu \sim T^2$. The first correction to the magnetization $\Delta M_+ \sim (\Delta T \mu)^2$ because according to the equation $M'(\delta \mu_{ex}) = 0$ and the correction to the envelope of magnetization

$$\Delta M_+ = M'(\delta \mu_{ex}) - M(\delta \mu_{ex}) = M'(\delta \mu_{ex}) \Delta T \mu_{ex} + \frac{d^2 M(\delta \mu_{ex})}{d(\delta \mu_{ex})^2} \frac{(\Delta T \mu_{ex})^2}{2} =$$

$$= -\frac{d^2 M(\delta \mu_{ex})}{d(\delta \mu_{ex})^2} \frac{(\Delta T \mu_{ex})^2}{2} = C^* \frac{d^2 \tilde{n}_0(\delta \mu_{ex}^0)}{d(\delta \mu_{ex}^0)^2} \frac{(\Delta T \mu_{ex})^2}{2} \quad (38)$$

In writing this we have used $M'(\delta \mu_{ex}^0) = M'(\delta \mu_{ex}) - M''(\delta \mu_{ex}) \Delta T \mu_{ex}$. From (34) we have
\[ \hat{n}_0' (\delta \mu_{ex}) \approx D_0' (\delta \mu_{ex}) \] (39)

In our case (32)

\[ D_0' (\delta \mu_{ex}) = -\frac{\sinh \left( \frac{\delta \mu_{ex}}{2\Gamma} \right)}{4\Gamma^2 \cosh \left( \frac{\delta \mu_{ex}}{2\Gamma} \right)} \approx \frac{1}{\Gamma \hbar \omega_c} , \]

\[ D_0''(\delta \mu_{ex}) \approx \frac{3}{2\Gamma^3 \hbar \omega_c} \quad \text{and} \quad \Delta T \mu_{ex} = -\frac{T^2 \pi^2}{4 \Gamma} \]

Substituting this into (38) and (39) and using (35) we obtain the envelope in the first approximation

\[ M^1_{\pm} = C^* \left[ \frac{1}{2} - \frac{\Gamma}{\hbar \omega_c} \ln \left( \frac{\hbar \omega_c}{\Gamma} \right) - \frac{\Gamma}{\hbar \omega_c} - \frac{1}{2} \left( \frac{T}{\Gamma} \right)^2 \right] \] (40)

B. Lorentzian shape of LLs

For the Lorentzian shape of LLs

\[ D_0(E) = \frac{1}{\pi \Gamma} \frac{1}{1 + (E/\Gamma)^2} \] (41)

we shall do the same steps as for the exponentially falling \( D(E) \). From equation (29) in zeroth approximation we obtain

\[ \delta \mu_{ex} = \Gamma \sqrt{\frac{\hbar \omega_c}{\pi \Gamma}} - 1 \approx \sqrt{\Gamma \hbar \omega_c / \pi} \] (42)

and

\[ \hat{n}_0(\delta \mu_{ex}) = \frac{1}{\pi} \arctan \left( \frac{\delta \mu_{ex}}{\Gamma} \right) + \frac{1}{2} \] (43)

Substituting this into the expression for magnetization (23) we get

\[ M^0_{\pm} = \pm C^* \left[ \frac{1}{2} - 2 \sqrt{\frac{\Gamma}{\pi \hbar \omega_c}} \right] \] (44)

This is different from (33). So, the envelope is different for different shapes of LLs. The temperature correction is also different.

To obtain the first temperature correction we need

\[ D_0'(\delta \mu_{ex}) \approx \frac{2}{\hbar \omega_c} \sqrt{\frac{\pi}{\Gamma \hbar \omega_c}} \quad \text{and} \quad D_0''(\delta \mu_{ex}) \approx \frac{6\pi}{\Gamma (\hbar \omega_c)^2} \]

\[ \Rightarrow \Delta T \mu_{ex} = -\frac{\pi^{5/2} T^2}{2 \sqrt{\Gamma \hbar \omega_c}} \]

Substituting this into (38) and (23) and using (44) we get

\[ M^1_{\pm} = C^* \left[ \frac{1}{2} - 2 \sqrt{\frac{1}{\pi \hbar \omega_c}} - 2 \sqrt{\frac{1}{\pi \hbar \omega_c} \cdot \pi^4 T^4} \right] \] (45)

The formulas (40) and (45) are not very confident even at low temperature. This is because the oscillating part of the DoS distribution function depends not only on the one parameter – the width \( \Gamma \),
but also on the LL separation $\hbar\omega_c$ and hence on the magnetic field. Any case the approximate functions \textcolor{red}{2} and \textcolor{red}{11} may be too far from reality. The exponentially falling $D_0(E)$ is always the incorrect approximation, while the confidence of the Lorentzian shape of LLs in 2D metals will be discussed later. The envelope of magnetization oscillations is very sensitive to the tails of DoS function $D(E)$ at low temperature and one should use more realistic (probably magnetic field dependent) functions $D(E)$ for accurate calculations.

Nevertheless, the formulas \textcolor{red}{10} and \textcolor{red}{13} can be used to indicate the qualitative features. They predict that the first temperature correction to the envelope is proportional to $T^4$ and that this envelope depends strongly on the shape of LLs at low temperature.

We can now say also at what conditions the previous results are valid. At $N = \text{const}$ and finite temperature the only results \textcolor{red}{1}, \textcolor{red}{3} for magnetization and its envelope have been obtained for sharp LLs ($D(E) = \delta(E)$). These formulas have been derived also in the limit $T \ll \hbar\omega_c$. If we assume the shape of LLs to be Lorentzian these result are valid if $\hbar\omega_c \gg T \gg \sqrt{\hbar\omega_c}$. This narrow region may not exist at all, that explains why many experimental data cannot be described by these formulas.

V. MAGNETIZATION OSCILLATIONS AT CONSTANT CHEMICAL POTENTIAL

The case of constant chemical potential may take place in organic metals when the electron reservoir formed by the one-dimensional parts of FS is very large. This has been observed recently on $\beta''$-(BEDT-TTF)$_2$SF$_6$$_\text{CH}_2$CF$_2$SO$_3$. \textcolor{red}{14} by the inverse sawtooth waveform of the magnetization oscillations.

The case $\mu = \text{const}$ may take place also due to giant magnetostriction (volume change) that cancels the variations of chemical potential associated with the oscillations of the density of states. This has been observed in beryllium \textcolor{red}{3}. In organic metals the electron density is not usually large enough to observe the giant magnetostriction. The restriction $\mu = \text{const}$ can be achieved, in principle, by connecting the sample with some big 3D piece of metal.

The case $\mu = \text{const}$ is interesting because it is more favorable to observe the diamagnetic phase transition in 2D metal and Condon domains \textcolor{red}{21}. It is also interesting to compare the cases $\mu = \text{const}$ and $N = \text{const}$.

Although in pure 2D materials at low temperature the dHvA oscillations are essentially non sinusoidal, the expansion over harmonics is still useful and the infinite harmonic series for some special densities of states can be calculated with good accuracy. The 2D harmonic expansion of the magnetization oscillations has been first obtained by Shoenberg \textcolor{red}{6} in the case $\mu = \text{const}$ using the intuitive phase smearing arguments. In the special case of the Lorentzian shape of LLs it has been derived recently \textcolor{red}{7} using Green’s function. We shall give another simple derivation of this harmonic expansion for arbitrary shape of LLs. By definition the thermodynamic potential is given by the same expressions \textcolor{red}{8} as in the case $N = \text{const}$. We assume $D(x)$ to be independent of the number of a LL, since in dHvA effect only the LLs near FS are important and for these levels the function $D(x)$ does not differ substantially. Then $\rho(E) = \rho(E/\hbar\omega_c)$ is a periodic function. One can take the spin-splitting into account by introducing $\rho_s(E) = \rho(E/\hbar\omega_c - \Delta/2) + \rho(E/\hbar\omega_c + \Delta/2)$, where $\Delta$ is the energy of spin splitting divided by $\hbar\omega_c$. This is equivalent to introducing $\Omega_s(\mu) = \Omega(\frac{\hbar\omega_c}{\mu} - \Delta/2) + \Omega(\frac{\hbar\omega_c}{\mu} + \Delta/2)$, since integration in \textcolor{red}{8} is between infinite limits. Spin will be recovered in the final result by introducing $M_s(\mu) = M(\frac{\hbar\omega_c}{\mu} - \Delta/2) + M(\frac{\hbar\omega_c}{\mu} + \Delta/2)$, where $M(\mu)$ is the magnetization without spin.

The density of states \textcolor{red}{1} can be rewritten as a harmonic series

$$\rho(E, B) = \frac{g}{\hbar\omega_c} \sum_{k=\infty}^{\infty} \exp \left[ 2\pi i \left( \frac{E}{\hbar\omega_c} - \frac{1}{2} \right) k \right] A(k) \frac{1 + \text{sign}(E)}{2}$$

where we introduced the Fourier transform of the DoS distribution on each LL

$$A(k) = \int_{-\frac{\hbar\omega_c}{2}}^{\frac{\hbar\omega_c}{2}} D(E') \exp \left( 2\pi i \frac{E'k}{\hbar\omega_c} \right) dE'$$

and the factor $(1 + \text{sign}(E))/2$ indicates that no electron states exist for $E < 0$. One can omit the term with $k = 0$ since it doesn’t affect the magnetic oscillations. Substituting \textcolor{red}{46} into \textcolor{red}{1} and integrating two times by parts we get
\[ \hat{\Omega} = -\frac{g}{\hbar \omega_c} \int \frac{df}{dE} \sum_{k \neq 0} \frac{\hbar^2 \omega_c^2}{(2\pi k)^2} \exp \left[ 2\pi i \left( \frac{E}{\hbar \omega_c} - \frac{1}{2} \right) k \right] A(k) \] (48)

Since \( \frac{\partial f}{\partial E} \neq 0 \) only in a small region near FS, the factor \( (1 + \text{sign}(E))/2 \) has been omitted.

Introducing the Furrier transform of \( \frac{\partial f}{\partial E} \)

\[ F_T(k) = \int dE \left( -\frac{\partial f}{\partial E} \right) \exp \left( 2\pi i \left( \frac{E - \mu}{\hbar \omega_c} \right) k \right) = \frac{2\pi^2 Tk/\hbar \omega_c}{\sinh(2\pi^2 Tk/\hbar \omega_c)} = \frac{k \lambda}{\sinh k \lambda} \] (49)

we get

\[ \hat{\Omega} = \frac{g \hbar \omega_c}{2\pi^2} \sum_{k=1}^{\infty} \frac{(-1)^k}{k^2} \cos \left( \frac{\mu}{\hbar \omega_c} 2\pi k \right) A(k) F_T(k) \] (50)

Differentiating (50) we obtain the harmonic expansion of the magnetization oscillations

\[ \hat{M} = -\frac{\partial \hat{\Omega}}{\partial B} \bigg|_{\mu=\text{const}} = \frac{g \mu}{\pi B} \sum_{k=1}^{\infty} \frac{(-1)^{k+1}}{k} \sin \left( \frac{\mu}{\hbar \omega_c} 2\pi k \right) A(k) F_T(k) \] (51)

This coincides with the Shoenberg's formula [3].

The oscillations of the electron density are given by

\[ \hat{N} = -\frac{\partial \hat{\Omega}}{\partial \mu} \bigg|_{B=\text{const}} = \frac{g}{\pi} \sum_{k=1}^{\infty} \frac{(-1)^k}{k} \sin \left( \frac{\mu}{\hbar \omega_c} 2\pi k \right) A(k) F_T(k) \] (52)

From (51) and (52) the simple relation follows

\[ \hat{M} = -\mu \hat{N}/B. \] (53)

It is valid only in the limit \( \mu \gg \hbar \omega_c \) where the formula (51) is valid.

In the case \( N = \text{const} \) one can obtain the similar relation between magnetization and the oscillations of the chemical potential. The magnetization oscillations at \( N = \text{const} \)

\[ \hat{M} = -\frac{d(\hat{\Omega} + N \mu)}{dB} \bigg|_{N=\text{const}} = -\frac{\partial \hat{\Omega}}{\partial \mu} \bigg|_{\mu, N=\text{const}} - \]

\[ - \left( \frac{\partial \hat{\Omega}}{\partial \mu} \bigg|_{N, B=\text{const}} + N \right) \frac{d\mu}{dB} \bigg|_{N=\text{const}} = -\frac{\partial \hat{\Omega}}{\partial \mu} \bigg|_{\mu, N=\text{const}} \]

So, the formula (51) is valid also in the case \( N = \text{const} \) but the chemical potential becomes an oscillating function of the magnetic field. The total electron density

\[ N = \int_{-\infty}^{\mu} [n_0(E) + n_R(E)]dE + \hat{N} = \text{const} \] (54)

where \( n_0(E) = (g/\hbar \omega_c) \theta(E) \) is the average DoS on the 2D part of FS and \( n_R(E) \) is the reservoir DoS. Substituting \( N = \int_{-\infty}^{E_F} [n_0(E) + n_R(E)]dE \) and (53) we get

\[ \hat{M} = \frac{\mu}{B} \int_{E_F}^{\mu} [n_0(E) + n_R(E)]dE \] (55)

If \( n_R(E) = \text{const} \) this simplifies

\[ \hat{M} = \frac{\mu}{B} (n_0 + n_R)(\mu - E_F). \] (56)

This expression coincides with the formula (3). The variations of the chemical potential \( |\mu - E_F| < \hbar \omega_c/2 \) and the relation (54) is valid if \( n_R(E_F) \hbar \omega_c/2 \ll n_R(E_F) \). This is usually fulfilled in the limit \( \mu/\hbar \omega_c \gg 1 \).
The formula (56) has been obtained previously only for sharp LLs in \cite{9} and for Lorentzian shape of LLs without reservoir states in \cite{6}. We have derived this formula in general case so that its applicability region becomes clear.

The electron reservoir (the independent of the magnetic field DoS) does not contribute to the magnetization oscillations at $\mu = const$. This is evident from (6) since $\Omega$ is linear in $\rho(E, B)$ and the DoS $\rho(E, B)$ is the only magnetic field dependent factor in right hand of the expression (6). So, in the case $\mu = const$ one can consider only oscillating part of the DoS.

The expression (51) depends essentially on the function $A(k)$ that is determined by shape of LLs. The question about the DoS distribution on the LLs in 2D metals is open. The Lorentzian shape of LLs (41) at small LL broadening $\Gamma$ leads to the Dingle low of harmonic damping

$$A(k) = \exp\left(\frac{\Gamma k}{\hbar\omega_c}\right)$$

This exponential low of harmonic damping has a direct physical meaning – it assumes the noncoherent scattering of the electrons on the impurities. The width of LLs $\Gamma$ is related to the electron scattering relaxation time $\tau = \pi\hbar/\Gamma$ and (57) is the amplitude of the probability that an electron return to the initial state after $k$ cyclotron periods. This can be shown by follows. The DoS is connected with the electron Green’s function by the relation

$$\rho(E) = -\frac{1}{\pi}ImG^R(r, r, E)$$

The imaginary part of the retarded Green’s function

$$ImG^R(r, r, E) = \frac{1}{2}(G^R - G^A) = -\frac{1}{2}\int_{-\infty}^{\infty} i < \psi^+(r, 0)|\psi(r, t) + \psi(r, t)|\psi^+(r, 0) > \exp(-iEt/\hbar)dt$$

Substituting this into (47) we get

$$A(k) = \int_{-\infty}^{\infty} \rho(E) \exp\left(\frac{2\pi iEk}{\hbar\omega_c}\right) dE =$$

$$= \frac{1}{2\pi} \int_{-\infty}^{\infty} dt < \psi^+(r, 0)|\psi(r, t) + \psi(r, t)|\psi^+(r, 0) > \int_{-\infty}^{\infty} dE \exp\left(\frac{2\pi iEk}{\hbar\omega_c} - \frac{iEt}{\hbar}\right) dE =$$

using

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} dE \exp\left(\frac{2\pi iEk}{\hbar\omega_c} - \frac{iEt}{\hbar}\right) dE = \delta(t - 2\pi k/\omega_c)$$

we finely obtain

$$A(k) = < \psi^+(r, 0)|\psi(r, T_c k) + \psi(r, T_c k)|\psi^+(r, 0) >$$

where $T_c = 2\pi/\omega_c$ is the cyclotron period.

The formula (53) states for the arbitrary ratio $\Gamma/\hbar\omega_c$ and is more general than (41). In 3D metals after each scattering process an electrons leave the impurity for a long time because it is free in the direction of the magnetic field. In 2D case the electrons can not move in $z$ direction and return to the same impurity after each cyclotron period. If the phase smearing time is longer than the cyclotron period the electron interfere with itself and this may change the amplitude (57) and hence the shape of LLs. So, the Dingle low of harmonic damping may violate if the cyclotron energy is greater than warping and the width of LLs $\hbar\omega_c \gg W, \Gamma$. Nevertheless on many experiments on organic metals no remarkable deviation from the Dingle law of harmonic damping has been observed. This may be because even small $k_z$ dispersion breaks the coherence of 2D scattering. We now assume the Dingle low (57) to be valid and try to sum the series (51) for magnetization oscillations at arbitrary $T$ and $\Gamma$.

The oscillating part of the magnetization is
\[ \tilde{M} = \frac{g\mu}{\pi B} \text{Im} \sum_{k=1}^{\infty} \exp \left[ i y k - b k \right] \left( -1 \right)^{k+1} \frac{\lambda}{\sinh k\lambda} \] (61)

where

\[ \lambda = 2\pi^2 T/\hbar\omega, \quad b = \pi/\omega_c \tau \quad \text{and} \quad y = \frac{2\pi\mu}{\hbar\omega_c} \] (62)

To calculate this sum we expand

\[ \frac{1}{\sinh(k\lambda)} = \frac{2e^{-k\lambda}}{1 - e^{-2k\lambda}} = \sum_{p=0}^{\infty} 2e^{-k\lambda} e^{-2pk\lambda} \] (63)

Now sum over \( k \) is the sum of geometric series and can be calculated

\[ \tilde{M} = \frac{2\mu\lambda}{\pi B} \text{Im} \sum_{p=0}^{\infty} Q(p) \] (64)

where

\[ Q(p) = \frac{\exp \left[ iy - b + \lambda(1 + 2p) \right]}{1 + \exp \left[ iy - b - \lambda(1 + 2p) \right]} \] (65)

\( Q(p) \) is a smooth monotonic function so one can apply the Euler-Maclaurin summation formula \[19\]

\[ \sum_{p=0}^{\infty} Q(p) = \frac{1}{2} Q(0) + \int_0^{\infty} Q(p) dp - \sum_{j=1}^{\infty} \frac{Q^{(2j-1)}(0)}{(2j)!} B_{2j} \] (66)

where \( B_k \) are the Bernoulli numbers. The terms of the sum in the right-hand part of eq. (66) fall off \( \sim \frac{(2\lambda)^k}{(k+1)!} \), hence their sum converge rapidly. The first term \( \sim \frac{\lambda}{4} Q(0) \). So, all the sum in the right-hand part is much less than \( Q(0) \) and we can ignore it. The integral

\[ \text{Im} \int_0^{\infty} Q(p) dp = \frac{1}{2\lambda} \arctan \left( \frac{\sin(y) \exp[-b - \lambda]}{1 + \cos(y) \exp[-b - \lambda]} \right) \] (67)

Substituting (63), (66) and (67) into (64) and recovering the spin splitting we obtain

\[ \tilde{M}_s = \frac{\mu}{\pi \Phi_0} \arctan \left( \frac{\sin(y - \pi \Delta) \exp[-b - \lambda]}{1 + \cos(y - \pi \Delta) \exp[-b - \lambda]} \right) + \lambda \sin(y - \pi \Delta) \exp[-b - \lambda] + \frac{\lambda \sin(y - \pi \Delta) \exp[-b - \lambda]}{1 + 2 \cos(y - \pi \Delta) \exp[-b - \lambda] + \exp[-2b - 2\lambda]} \] (68)

This expression gives the amplitude and the shape of magnetization oscillations. It is much more convenient for practical use in the limit of weak harmonic damping than the infinite series (51). In the limit of strong harmonic damping \( \lambda \gg 1 \) or \( b \gg 1 \) formula (68) correctly gives the first harmonics. In the other limit \( \lambda = 0 \) the formula (68) becomes exact and simple

\[ \tilde{M}_s = \frac{\mu}{\pi \Phi_0} \arctan \left( \frac{\sin(y - \pi \Delta) e^{-b}}{1 + \cos(y - \pi \Delta) e^{-b}} \right) + \{ (y - \pi \Delta) \rightarrow (y + \pi \Delta) \} \]

This coincides with the recent result of T. Champel and V.P. Mineev \[3\]. At \( b = 0 \), using the identity

\[ \arctan \left( \frac{\sin x}{1 + \cos x} \right) = \arctan \left( \frac{\sin(x/2)}{\cos(x/2)} \right) \]

one gets a saw-tooth function as has been predicted by Peierls \[20\].
If spin splitting energy is just n-times the cyclotron energy (this can be achieved by tilting the magnetic field) and chemical potential $\mu \gg \hbar \omega_c$, formula (68) substantially simplifies

$$M_0 = \frac{2g\mu}{\pi B} \left[ \arctan \left( \frac{\sin y \exp[-b - \lambda]}{1 + \cos y \exp[-b - \lambda]} \right) + \frac{\lambda \sin y \exp[-b - \lambda]}{1 + 2 \cos y \exp[-b - \lambda] + \exp[-2b - 2\lambda]} \right]$$

Now one can obtain the envelope of magnetization oscillations. The equation $\frac{\partial M}{\partial B} = 0$ gives

$$\cos y^* = \frac{-(1 + 3ex^2 + \lambda + \lambda ex^2) + \sqrt{(1 + 3ex^2 + \lambda + \lambda ex^2)^2 - 8ex^2(1 + 2\lambda + ex^2)}}{4ex}$$

where $ex \equiv \exp[-b - \lambda]$, that determines the positions of extrema of $M(B)$. Substitution this into (69) gives the envelope of magnetization oscillations. In the limit $\lambda \ll 1$, $b \ll 1$, $\lambda \sim b$ we have

$$M_{\pm}(B) \simeq \pm \frac{\mu S}{\Phi_0} \left[ 1 - \frac{2\sqrt{2}b}{\pi} + O \left( \lambda^2, b^2 \right) \right]$$

where $\lambda$ and $b$ are given by (62). The result $M_{\pm}$ does not depend on temperature in first approximation at $\lambda \sim b \ll 1$. This is not surprising because $A(k) = \exp(-bk)$ has stronger dependence on $b$ than $F_r(k)$ (eq. [49]) on $\lambda$ for small $k$, that are important in the sum (51). Since $b = 2\pi T/\hbar \omega_c$ the formula (71) gives the same envelope as (51). Thus, if the Dingle low of harmonic damping is valid, the envelope of magnetization oscillations is identical in the cases of constant particle density and fixed chemical potential in the limit $b = 2\pi T/\hbar \omega_c \ll 1$ and zero temperature.

Using formula (58) one can examine the possibility of the diamagnetic phase transition (DPT) [5]. It must take place if $dM/dB > 1/4\pi$. It has been observed in some pure 3D metals due to the formation of Condon domains [21]. In 2D case it can lead to an energy gap at the FS, analogies to that of QHE. The diamagnetic phase transition is not possible in available heterostructures since the magnetization is $\sim 5$ orders less than necessary. In organic metals the electron density is much greater than in heterostructures and the constraint $\mu = const$ is more favorable to observe the DPT. For example, in the compound $\kappa = (BEDT - TTF)_2I_3$ at $T_D = 0.1K$ the slope $dM/dB$ turns out to be only 5 times smaller than needed for this phase transition. Maybe there exist another compound where this derivative $dM/dB$ is great enough.

There is another characteristic parameter of magnetization oscillations, besides the frequency and the amplitude, that can be compared with the experiment. It is the value of the magnetic field $B_0$ above which the spin-splitting is clearly visible on the graph $M(B)$. More formally, it is the point $B_0$, above which the first derivative of magnetization has an additional zero on each period due to the spin-splitting. This point $B_0$ is extremely sensitive to the parameters of the system, such as the tilting angle of the magnetic field, the shape of LLs and the reservoir DoS, the temperature and so on. But to obtain an accurate theoretical description of this critical field one needs to make the numerical calculations to take into account many factors, unimportant for the amplitude of the oscillations.

In the 2D organic metals the magnetic breakdown [1] usually takes place at some field $B_{MB}$. The frequency of dHvA oscillations is different below and above $B_{MB}$. In the vicinity of $B_{MB}$ one should apply more complicated theory of the dHvA effect (21) and the references therein). But far from $B_{MB}$ all formulas remain valid, but one should always substitute $\mu$ by $\hbar \omega_c \cdot F/B$, where $F$ is the new dHvA frequency.

VI. CONCLUSION

The dHvA effect in normal 2D metals has been investigated at different conditions and at the arbitrary shape and width of LLs. Finite temperature, spin-splitting and electron reservoir states are taken into account. In the limit $E_F/\hbar \omega_c \gg 1$ the formulas greatly simplifies and many results can be obtained analytically. In this limit there are simple relations (53) and (56) between the magnetization oscillations and the oscillations of the chemical potential (in the case $N = const$) or the oscillations of the electron density (in the case $\mu = const$). These relations are valid at the arbitrary shape of LLs.

We have obtained the analytical formulas for the magnetization and its envelope at different conditions and shapes of LLs, that may be useful to treat the experimental data. The envelope of the magnetization
oscillations depends strongly on the shape of LLs in the limit of weak harmonic damping (compare the formulas 40 and 45).

We derived the explicit relation between the wave form of the magnetization oscillations and the shape of LLs at arbitrary reservoir DoS. This relation makes possible to obtain the DoS distribution at arbitrary magnetic field and spin-splitting from the measurement of the dHvA effect. The DoS distribution gives much more information about the electronic structure of compounds than the Dingle temperature and the effective electron mass. This may help to solve the theoretical problem about the DoS distribution in 2D metals in magnetic field.

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