

THE DE HAAS - VAN ALPHEN EFFECT IN QUASI-TWO-DIMENSIONAL MATERIALS

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We calculate the amplitude of magnetization oscillations for quasi-two-dimensional electron system. In two-dimensional case the behavior of this amplitude as a function of magnetic field and temperature differ completely from the conventional Lifshitz-Kosevich formula valid for 3-dimensional metals. Before only ideal two-dimensional case has been considered, and the difference of the shape of Fermi surface from cylindrical one has not been taken into account. We obtain the general formula for the envelope of magnetization oscillations as a function of magnetic field, temperature and the strength of the warping of Fermi surface. This problem is important because of the aroused in recent years a great interest to heterostructures and quasi-2D organic metals.

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1. Introduction. The de Haas - van Alphen (dHvA) effect is very important for investigating the properties of metals, since it gives the information about Fermi surface and cyclotron mass of electrons in these compounds. This effect has been suggested in 1930 by L.D.Landau [1] and has been first measured by de Haas and van Alphen in the same year [2]. The calculation of magnetization as a function of magnetic field and temperature for arbitrary electronic spectrum in 3-dimensional metals has been carried out in 1955 by Lifshitz and Kosevich [3]. These authors assume the chemical potential to be constant, that is a very good approximation in 3-dimensional case but not valid for two dimensions. This difference takes place because in three dimensions the spectrum of electrons is continuous (due to z -component of momentum) and the Fermi surface is intersected by many Landau levels(LLs). Therefore the chemical potential is approximately equal to the Fermi energy and does not oscillate with changing of the magnetic field. In 2-dimensional(2D) system the electron spectrum has gaps(between LLs) and the chemical potential (which is the minimal energy of a particle to be added to the system) is pinned to the highest occupied Landau level(LL). When this LL becomes completely filled, chemical potential jumps to the next LL. Such a jump has been detected by the magnetization measurement in several single-layer, high-mobility 2D electron gas AlGaAs-GaAs heterostructures [4]. So, the chemical potential in two dimensions oscillates strongly with varying of magnetic field and must not be considered as a constant. This fact changes the amplitude and the form of magnetization oscillations completely.

On the experiment the significant departure from the conventional Lifshitz-Kosevich formula [3] has been observed in the number of the charge-transfer salts based on the molecule bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF) [5 - 7] and on the heterostructures

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[4]. The theoretical study of dHvA oscillations in ideal 2D case (with cylindrical Fermi-surface and without Landau level broadening) has been performed by Vagner et al.[8] in order to explain dHvA measurements made on 2D GaAs/Al_xGa_{1-x}As heterojunctions and graphite intercalation compounds. Analytical study of 2D dHvA effect with broadened Landau levels(LL) for zero temperature ($T = 0$)[9] and quite thorough numerical calculations[10] for a wide temperature region have been made, but in these papers the influence of warping of the Fermi-surface on the magnetization oscillations is disregarded. This approximation is correct only if the size of warping is much smaller than LL broadening and temperature. In this paper we consider the inverse case when this warping energy W is bigger than scattering broadening of LLs, but less (not much less!) than the distance between LLs. The analytical formula for the amplitude of magnetization oscillations is derived for arbitrary ratio of $W/2kT$. In the magnetic field, warping energy and temperature dependence of the amplitude of dHvA effect the information about warping of Fermi-surface and the effective mass is contained, so our formula will be very useful for obtaining these parameters from the experiment for many quasi-2D compounds.

2. Chemical potential. In order to calculate magnetization we need first to obtain the expression for chemical potential $\mu(B)$. Let us write the number of particles as a sum over all LL with Fermi distribution function f_n :

$$N = \frac{ga}{2\pi\hbar} \sum_{n=0}^{\infty} \int_0^{\frac{2\pi\hbar}{a}} f_n dp_z, \quad (1)$$

where g is the degeneracy of a LL, a is the lattice constant in z direction (the 2D layer separation in heterostructures) and N – is the number of particles in one layer. The chemical potential is situated between the two LLs with energies $\hbar\omega_c(n_F - \frac{1}{2})$ and $\hbar\omega_c(n_F + \frac{1}{2})$ where $\omega_c = eB/m^*c$ – is the cyclotron frequency. Since we assume $\hbar\omega_c \gg kT$, only these two Landau levels make contribution to thermodynamics. So, we can set $f_n = 1$ for all $n < n_F - 1$ and $f_n = 0$ for all $n > n_F$:

$$N = (n_F - 1)g + \frac{ga}{2\pi\hbar} \int_0^{\frac{2\pi\hbar}{a}} (f_{n_F-1} + f_{n_F}) dp_z. \quad (2)$$

The energy of electron is given by $E = \hbar\omega_c(n + \frac{1}{2}) + E_z$, where n is the number of LL and the term $E_z = (W/2)(1 - \cos(p_z a/\hbar))$ gives the dependence of energy on the momentum along z -axis. For ideal 2D case $W = 0$, and $W \neq 0$ takes into account the warping of Fermi surface. Later for convenience we shall use another expression for E_z :

$$E_z = (W/2) \cos(p_z a/\hbar). \quad (3)$$

The difference between expression (3) and the previous one consists of only shifting the starting point of energy by a constant $W/2$ (which of course does not change any physical result) and the shifting of starting point of quasi-momentum p_z by $\pi\hbar/a$ (that makes no difference because of subsequent integration over the full period of p_z). Substituting the expression for $f_n = 1/[1 + \exp(E - \mu/kT)]$ into eq. (2) we get

$$\begin{aligned} \frac{N}{g} - (n_F - 1) &= \frac{1}{2\pi} \int_0^{2\pi} \frac{dy}{1 + \exp(X_F - \alpha) \exp[(W/2kT) \cos y]} + \\ &+ \frac{1}{2\pi} \int_0^{2\pi} \frac{dy}{1 + \exp(X_F + \alpha) \exp[(W/2kT) \cos y]}, \quad (4) \end{aligned}$$

where $y = p_z a / \hbar$, $X_F = (\hbar\omega_c n_F - \mu) / kT$, and $\alpha = \hbar\omega_c / 2kT$. In order to calculate the integral we will use the condition

$$\exp(-\alpha + W/2kT \pm X_F) \ll 1 \quad (5)$$

which as will be shown later even for $W \sim \hbar\omega$ is equivalent to $\sqrt{\alpha} \gg 1$ and is usually fulfilled in the experiments. So, one can make an expansion of (4) over small parameter (5) keeping only first two terms:

$$\begin{aligned} \frac{N}{g} - (n_F - 1) &= \frac{1}{2\pi} \int_0^{2\pi} [1 - e^{X_F - \alpha} \exp\left(\frac{W}{2kT} \cos y\right) + e^{-X_F - \alpha} \exp\left(\frac{-W}{2kT} \cos y\right)] dy = \\ &= 1 - e^{-\alpha} \cdot 2 \sinh X_F \cdot I_0\left(\frac{W}{2kT}\right), \end{aligned} \quad (6)$$

where $I_0(W/2kT)$ is the modified Bessel function of argument $W/2kT$. Now we have

$$N/g - n_F = \tilde{n} = -e^{-\alpha} \cdot 2 \sinh X_F \cdot I_0(W/2kT). \quad (7)$$

Later we shall use the notation $\tilde{n} = N/g - n_F$. Equation (7) can be easily solved with respect to chemical potential:

$$\sinh X_F = -\frac{\tilde{n} e^\alpha}{2I_0\left(\frac{W}{2kT}\right)} \Rightarrow \mu = \hbar\omega_c n_F + kT \operatorname{arsh} \left(\frac{\tilde{n} e^\alpha}{2I_0\left(\frac{W}{2kT}\right)} \right). \quad (8)$$

Let us see the limiting cases of the obtained expression for μ . For this purpose we need the asymptotic behavior of modified Bessel function:

$$I_0(x) = 1 + \frac{x^2}{4} + \dots, \quad x \ll 1; \quad (9)$$

$$I_0(x) = \exp(x) \cdot \sqrt{1/2\pi x}, \quad x \gg 1. \quad (10)$$

For $W/2kT \ll 1$, formula (8) coincides with the expression for chemical potential without warping of Fermi surface [8] $\mu = \hbar\omega_c n_F - kT \ln[(-\tilde{n} \cosh \alpha + \sqrt{1 + \tilde{n}^2 \sinh^2 \alpha}) / (1 + \tilde{n})]$. To show this we'll simplify this expression using $\tilde{n} e^\alpha \gg 1$: $\mu \simeq \hbar\omega_c n_F - kT \ln(\tilde{n} e^\alpha / (1 - \tilde{n}))$. Taking into account identity $\operatorname{arsh} x = \ln(x + \sqrt{1 + x^2})$ and inequality $\tilde{n} e^\alpha \gg 1$ we can rewrite formula (8) in the form $\mu = \hbar\omega_c n_F - kT \ln(\tilde{n} e^\alpha)$. So, in the limit $\tilde{n} \ll 1/\sqrt{\alpha}$ these formulae coincide.

3. Thermodynamic potential and magnetization For calculating the thermodynamic potential we'll use its definition:

$$\Omega = -kT \frac{g a}{2\pi \hbar} \int_0^{2\pi \hbar} \sum_{n=0}^{\infty} \ln \left(1 + \exp \left[\frac{\mu - E_n(p_z)}{kT} \right] \right) dp_z. \quad (11)$$

The calculation of sum and integral is analogous to made up for chemical potential.

Making the expansion over the same small parameter $\exp(-\alpha + W/2kT \pm X_F) \ll 1$ and separating the contribution from the last two LL we get:

$$\begin{aligned} \Omega = g \sum_{n=0}^{n_F-2} \left(\hbar\omega_c \left(n + \frac{1}{2} \right) - \mu \right) + \frac{gkT}{2\pi} \int_0^{2\pi} \left[(X_F - \alpha + \frac{W}{2kT} \cos y) - e^{X_F - \alpha} \times \right. \\ \left. \times \exp\left(\frac{W}{2kT} \cos y\right) \right] dy + \frac{gkT}{2\pi} \int_0^{2\pi} \left[-e^{-X_F - \alpha} \exp\left(\frac{-W}{2kT} \cos y\right) \right] dy. \end{aligned}$$

After taking the integrals and sum we obtain finely

$$\Omega = g[(n_F^2/2)\hbar\omega_c - \mu n_F] - gkTe^{-\alpha} \cdot 2 \cosh X_F \cdot I_0(W/2kT). \quad (12)$$

It is easy to check that in the limit $W/2kT \ll 1$ and $\bar{n} \ll 1$ this formula coincides with the expression for thermodynamic potential in the 2D case of ideally cylindric Fermi surface [8]

$$\Omega = g \left[\frac{n_F^2}{2} \hbar\omega_c - \mu n_F + \frac{1}{2} \hbar\omega_c - kT \ln(2 \cosh X_F + 2 \cosh \alpha) \right]$$

(for proving this one should use $\cosh X_F \simeq |\bar{n}| e^\alpha / 2I_0(W/2kT) \ll \cosh \alpha$). Free energy

$$F = \Omega + \mu N = g \left\{ \frac{n_F^2}{2} \hbar\omega_c + \left(\frac{N}{g} - n_F \right) \mu - kTe^{-\alpha} \cdot 2 \cosh X_F \cdot I_0 \left(\frac{W}{2kT} \right) \right\}. \quad (13)$$

Now we can calculate the magnetization

$$M = -\frac{\partial F}{\partial B} = -\frac{\partial F}{\partial \omega_c} \frac{e}{m^*c} - \frac{\partial F}{\partial g} \frac{S}{\Phi_0} - \frac{\partial F}{\partial \mu} \frac{\partial \mu}{\partial B} \quad (14)$$

(where S — is the area of the sample and Φ_0 — is the magnetic flux quantum). Now one only has to take this derivative accurately.

Since $g\hbar e/m^*c = \hbar\omega_c S/\Phi_0$ we get

$$M = \frac{S}{\Phi_0} \hbar\omega_c \left\{ -n_F \bar{n} + \frac{n_F}{2\alpha} \operatorname{arsh} \left(\frac{\bar{n} e^\alpha}{2I_0(W/2kT)} \right) + e^{-\alpha} \cdot I_0 \left(\frac{W}{2kT} \right) \cosh X_F \cdot \left(-1 + \frac{1}{\alpha} \right) \right\} \quad (15)$$

This is the expression for magnetization with warping of Fermi surface. The only restriction on the magnitude W of the warping is that it must be not greater than energy difference between LLs.

Formula (15) is valid only in the narrow region

$$\bar{n} \ll I_0(W/2kT) e^{-W/2kT} \simeq \min \left\{ \sqrt{kT/\pi W}, 1 \right\}, \quad (16)$$

where the condition (5): $\exp(-\alpha + W/2kT \pm X_F) \simeq \bar{n} e^{W/2kT} / I_0(W/2kT) \ll 1$ is satisfied. What we are interesting in is the envelope of magnetization oscillations, i.e. the amplitude of these oscillations as a function of magnetic field, temperature and the size of warping, because on the experiment only this amplitude is detected. To calculate this amplitude one need only the values of maxima and minima of magnetization oscillations. So if these extrema of magnetization get into these narrow regions of \bar{n} or B , then we can use formula (15) to calculate the envelope. As will be shown later, these extrema of $M(B)$ occur at $\bar{n} \simeq 1/2\alpha$, so that the condition (16) is usually satisfied (this restriction for $W \sim \hbar\omega_c$ is equivalent to $\sqrt{\alpha} \gg 1$).

Let us turn to the calculating of the envelope of $M(B)$. Since $|\sinh X_F| = |-\bar{n} e^\alpha / 2I_0(W/2kT)| \gg 1$, then $\cosh X_F \simeq |\sinh X_F| = |\bar{n}| e^\alpha / 2I_0(W/2kT)$, and the expression for magnetization (15) can be replaced by

$$M = \frac{SE_F}{\Phi_0} \left\{ -\bar{n} \left(1 + \frac{1}{n_F} \right) + \frac{1}{2\alpha} \operatorname{arsh} \left(\frac{\bar{n} e^\alpha}{2I_0(W/2kT)} \right) \right\}, \quad (17)$$

where $E_F = \hbar\omega_{c n_F}$ — is the Fermi energy of electrons without magnetic field. In order to find the amplitude of magnetization oscillations one has to differentiate (17) with respect

to B . Taking into account that $n_F(B) = \text{const}$ for each interval of B where the extrema occur, one get:

$$\frac{\partial M}{\partial B} \approx \frac{\hbar\omega_c}{B} \frac{gn_F}{B} \left\{ n_F + \frac{3}{2} - \frac{n_F}{2\alpha} \frac{1}{|\bar{n}|} \right\} = 0 \Rightarrow \bar{n}_{ex} = \pm \frac{1}{2\alpha} \frac{n_F}{n_F + 3/2}. \quad (18)$$

The values B_{ex} of magnetic field at which magnetization M has extrema are given by

$$B_{ex} = B^* \left(1 \pm \frac{1}{2\alpha(n_F + 3/2)} \right). \quad (19)$$

To obtain the envelope of magnetization one should substitute extremal values of \bar{n}_{ex} into (17) :

$$M_{\pm} = \pm \frac{SE_F}{2\Phi_0} \left\{ \frac{1}{\alpha} \text{arsh} \left(\frac{e^\alpha}{4\alpha I_0(W/2kT)} \frac{n_F}{n_F + 3/2} \right) - \frac{1}{\alpha} \frac{n_F + 1}{n_F + 3/2} \right\}. \quad (20)$$

All above calculations have been made for spinless electrons. For generalizing them one should substitute all $\hbar\omega_c$ in the resulting formula by the real distances between energy levels, that takes into account the spin splitting. These distances will depend periodically on the magnetic field; and this period in the energy scale is equal to LL separation. If spin-orbit interaction is weak compared to the cyclotron energy, the spin up states add one level between each two LL with spin down no matter what is the ratio of spin splitting energy to the distance between LLs (we assume that g -factor of electrons does not depend on the magnetic field). In the special cases, when the spin splitting energy is just n -times the distance between LLs (with the precision of the temperature or the LL broadening), the formula (20) acquires only the factor 2, because the effect of spin to magnetization oscillations in this case is only to increase the degeneracy of the LLs two times. Since temperature and LL broadening are not very small usually, such a situation is not rare. The envelope for magnetization oscillations in this case becomes:

$$M_{\pm} = \pm \frac{SE_F}{\Phi_0} \left\{ \frac{1}{\alpha} \text{arsh} \left(\frac{e^\alpha}{4\alpha I_0(W/2kT)} \frac{n_F}{n_F + 3/2} \right) - \frac{1}{\alpha} \frac{n_F + 1}{n_F + 3/2} \right\}. \quad (21)$$

In the limit $W/2kT \ll 1$ and $n_F \gg 1$ this formula coincides with previous one, obtained for the ideal two-dimensional case [8]

$$M_{\pm} = \pm \frac{SE_F}{\Phi_0} \left[1 - \frac{1}{\alpha} \ln(2\alpha) - \frac{1}{\alpha} \right], \quad (22)$$

which is valid when the number n_F of filled LL is large.

In the inverse case of large warping formula (20) becomes:

$$M_{\pm} = \pm \frac{SE_F}{\Phi_0} \left[1 - \frac{W}{\hbar\omega_c} + \frac{1}{\alpha} \ln \left(\frac{\sqrt{kT \cdot \pi W}}{\hbar\omega_c} \right) - \frac{1}{\alpha} \right]. \quad (23)$$

This formula differs substantially from the ideally-2D one (eq. (22)). The amplitude of the oscillations depends approximately linearly on the size of warping W .

4. Discussion. The obtained formula (eq. (20)) differs drastically from the 3D Lifshitz - Kosevich behavior. Instead of exponential dependence on temperature in 3D

case: $M \sim \exp(-2\pi^2 T c m^* / e \hbar H)$ the amplitude of magnetization oscillations has rather logarithmic dependence (eq. (23)). The form of the oscillations in two dimensions is also unsimilar to the 3D case. Instead of smooth nearly sinusoidal oscillations, where one can rest only few first harmonics in Lifshitz - Kosevich formula, in 2D case we have saw-tooth form. The maxima and minima of magnetization are separated by (see eq. (18)) $\Delta \bar{n} = n_F / \alpha (n_F + 3/2)$, so the jumps are very sharp if $\alpha \gg 1$, i.e. if temperature and LL broadening are much less than cyclotron energy. This result has an experimental confirmation[4]. So, considering only few first harmonics of the magnetization oscillations becomes bad approximation for obtaining the effective electron masses and other parameters of 2D compounds. Therefore using of the conventional Lifshitz-Kosevich formula for processing the experimental data for quasi-2D materials may lead to incorrect results.

For many BEDT-TTF salts, the effective mass m^* determined from the dHvA and Shubnikov - de Haas effects is found to be larger than the band mass m_b . This difference has been attributed to mass enhancement due to many-body renormalization [11]. Singleton et al.[12] have reported cyclotron resonance measurements indicating a cyclotron mass m_{CR} smaller by a factor of 5 than m^* on the closed orbit in the $\text{KHg}(\text{SCN})_4$ salt. They concluded that the transport mass is enhanced mainly by electron-electron interactions, whereas m_{CR} is independent of this short-range interactions, and the electron-phonon interactions may play only a minor role in the mass enhancement. Although the carrier on the relevant open orbit is thought to be also strongly correlated, direct evidence has not been obtained yet[13]. In this context it would be interesting to obtain the effective mass of these quasi-2D compounds from dHvA effect using our formula(20).

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