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THE INFLUENCE OF THE CHEMICAL POTENTIAL OSCILLATIONS ON THE DE HAAS–VAN ALPHEN EFFECT IN QUASI-TWO-DIMENSIONAL COMPOUNDS

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The de Haas–van Alphen effect in quasi-two-dimensional metals is studied at arbitrary parameters. Oscillations of the chemical potential can substantially change the temperature dependence of harmonic amplitudes that is commonly used to determine the effective electron mass. The processing of the experimental data using the standard Lifshitz–Kosevich formula can therefore lead to substantial errors even in the strong harmonic damping limit. This may explain the difference between the effective electron masses determined from the de Haas–van Alphen effect and the cyclotron resonance measurements. The oscillations of the chemical potential and the deviations from the Lifshitz–Kosevich formula depend on the reservoir density of states that exists in organic metals due to open sheets of the Fermi surface. This dependence can be used to determine the density of electron states on open sheets of the Fermi surface. We present analytical results of the calculations of harmonic amplitudes in some limiting cases that show the importance of the chemical potential oscillations. We also describe a simple algorithm for a numerical calculation of the harmonic amplitudes for arbitrary reservoir density of states, arbitrary warping, spin-splitting, temperature, and Dingle temperature.

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The quantum magnetization oscillations (or the de Haas–van Alphen (dHvA) effect) were discovered long ago [1] and have been widely used as a powerful tool in studying the Fermi surfaces and single electron properties in metals [2]. In a three-dimensional ($3D$) metal, a good quantitative description of this effect is given by the Lifshitz–Kosevich (LK) formula [3]. In two- or quasi-two-dimensional compounds, deviations from the LK formula are possible for three reasons: the harmonic damping in the two-dimensional ($2D$) case is different, the impurity scattering cannot be described by the usual Dingle law, and the chemical potential also becomes an oscillating function of the magnetic field. The first problem is important only when the harmonic damping is weak and can be easily solved using the $2D$ harmonic expansion [2]. The second problem concerns with an accurate calculation of the density of states (DoS) with electron–electron interactions and the impurity scattering. The electron–electron interactions are not very important if many Landau levels (LLs) are occupied (we consider the case where the Fermi energy ε_F is much greater than the Landau level separation and the temperature). In the $3D$ case, the impurity scattering adds an imaginary part $i\Gamma(B)$ to the electron spectrum, which means that the electron can leave its quantum state with the probability $w = \Gamma(B)/\pi\hbar$ per second. Assuming this energy level width $\Gamma(B)$ to be independent of the magnetic field B, one obtains the Dingle law of harmonic damping [4]

$$A_l \propto \exp\left(-2\pi l\Gamma/\hbar\omega_c\right),$$

where A_l is the amplitude of the harmonic number l and $\omega_c = eB/m^*c$ is the cyclotron frequency. This Dingle law has been proved by many experiments on $3D$ metals. In the $2D$ case, this law may be incorrect and the problem of the DoS distribution in $2D$ metals has not

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been solved yet, although many theoretical works have been devoted to this subject (for example, [5–7]). The problem is complicated because even the exact calculation of the point-like impurity scattering is not sufficient because the long-range impurities (and probably, the electron–electron interactions) are also important in the $2D$ case [8]. The procedure of extracting the DoS distribution from the dHvA measurements was recently proposed in [9]. In the present paper, we focus on the third question: we assume the Dingle law to be valid and consider the influence of the chemical potential oscillations on the harmonic amplitudes of the dHvA oscillations in this approximation. Because we consider the quasi-$2D$ case, the Dingle law is not a bad approximation. We show that the chemical potential oscillations substantially change the temperature and the Dingle temperature dependence of the harmonic amplitudes even in the limit of a strong harmonic damping. Therefore, the estimate of the effective electron mass based on the LK formula can lead to the error up to 30%. This can explain the difference between the effective electron masses obtained from the dHvA effect and from the cyclotron resonance measurements (for example, in [10] and [11]). This problem was examined numerically by Harrison et al. [12] at zero warping W of the Fermi surface (FS). In this paper, we derive explicit formulas describing the quantum magnetization oscillations at arbitrary parameters. We study the result analytically in some limiting cases. This shows the importance of the chemical potential oscillation effect on harmonic amplitudes.

The energy spectrum of the quasi-two-dimensional electron gas is given by

$$E_{n,k_z,\sigma} = \hbar\omega_c\left(n+\frac{1}{2}\right) + \frac{W}{2}\cos(k_z d) + \sigma\mu_e B, \quad (1)$$

where W is the warping of a quasi-cylindrical Fermi surface. The DoS distribution with the impurity scattering can be written as

$$\rho(E,B) = \rho_0(E,B) + \tilde{\rho}(E,B),$$

where for $E \gg \hbar\omega_c$, the oscillating part of the DoS is [13]

$$\tilde{\rho}(E,B) = \frac{4g}{\hbar\omega_c}\sum_{l=1}^{\infty}(-1)^l\cos\left(2\pi l\frac{E}{\hbar\omega_c}\right)\times \\ \times J_0\left(\pi l\frac{W}{\hbar\omega_c}\right)\cos\left(2\pi l\frac{\mu_e B}{\hbar\omega_c}\right)\exp\left(-\frac{2\pi l\Gamma}{\hbar\omega_c}\right). \quad (2)$$

In this formula, $g = B/\Phi_0$ is the LL degeneracy, the factor $\cos(2\pi l\mu_e B/\hbar\omega_c)$ is due to the spin splitting, and the factor $J_0(\pi lW/\hbar\omega_c)$ comes from the finite warping W of the quasi-cylindrical FS. $J_0(x)$ is the zero-order Bessel function. The last factor in (2) is the usual Dingle factor.

The non-oscillating part of the DoS is given by

$$\rho_0(E,B) = \frac{2g}{\hbar\omega_c}\left(1+n_R(E)\right),$$

where $n_R(E)$ is the ratio of the reservoir density of states to the average DoS on the quasi-$2D$ part of the FS. The reservoir density of states occurs in quasi-$2D$ organic metals because of open sheets of the FS. These quasi-one-dimensional states do not directly contribute to the magnetization oscillations because they form the continuous spectrum and the nonoscillationg DoS.

If the DoS is known, one can calculate the thermodynamic potential

$$\Omega(\mu,B,T) = \\ = -T\int_0^{\infty}\rho(E,B)\ln\left[1+\exp\left(\frac{\mu-E}{T}\right)\right]dE = \\ = \Omega_0(\mu,B,T) + \tilde{\Omega}(\mu,B,T), \quad (3)$$

where $\mu(B)$ is the chemical potential and the oscillating part of the thermodynamic potential is given by [13]

$$\tilde{\Omega} = 2gT\sum_{l=1}^{\infty}\frac{(-1)^l}{l}\cos\left(2\pi l\frac{\mu}{\hbar\omega_c}\right)\frac{\lambda l}{\mathrm{sh}(\lambda l)}\times \\ \times J_0\left(\pi l\frac{W}{\hbar\omega_c}\right)\cos\left(2\pi l\frac{\mu_e H}{\hbar\omega_c}\right)\exp\left(-\frac{2\pi l\Gamma}{\hbar\omega_c}\right),$$

where $\lambda \equiv 2\pi^2 T/\hbar\omega_c$. The total particle number is usually constant,

$$N = -\left(\frac{\partial\Omega(\mu,B,T)}{\partial\mu}\right)_{T,B} = \\ = \int_0^{\infty}\frac{\rho(E,B)}{1+\exp\left(\dfrac{E-\mu}{T}\right)}dE = \text{const}.$$

This is an equation for the chemical potential as a function of the magnetic field. Separating the oscillating part of the DoS and substituting

$$N = \int_0^{\infty}\frac{\rho_0(E,B)}{1+\exp\left(\dfrac{E-\varepsilon_F}{T}\right)}dE$$

(where ε_F is the Fermi energy at zero magnetic field), we obtain

$$\int_0^\infty \left(\frac{1}{1+\exp\left(\dfrac{E-\varepsilon_F}{T}\right)} - \frac{1}{1+\exp\left(\dfrac{E-\mu}{T}\right)} \right) \times$$
$$\times \rho_0(E,B)\,dE = \int_0^\infty \frac{\tilde{\rho}(E,B)}{1+\exp\left(\dfrac{E-\mu}{T}\right)}\,dE. \qquad (4)$$

We next use the fact that the reservoir DoS $n_R(E)$ does not change appreciably at the scale of T or $|\mu-\varepsilon_F| < \hbar\omega_c/2$ (this is true if many LLs are occupied because $n_R(E)$ changes substantially at the Fermi energy scale). It then follows that $n_R(E) \approx n_R(\varepsilon_F) = \mathrm{const} \equiv n_R$. The left-hand side of (4) can be simplified, and after the insertion of (2), we obtain the equation for the oscillating part $\tilde{\mu}(B)$ of the chemical potential,

$$\tilde{\mu}(B) \equiv \mu(B) - \varepsilon_F = \frac{\hbar\omega_c}{\pi(1+n_R(\varepsilon_F))} \times$$
$$\times \sum_{l=1}^{\infty} \frac{(-1)^{l+1}}{l} \sin\left(\frac{2\pi l\,(\varepsilon_F+\tilde{\mu}(B))}{\hbar\omega_c}\right) \frac{\lambda l}{\mathrm{sh}(\lambda l)} \times$$
$$\times \cos\left(2\pi l \frac{\mu_e H}{\hbar\omega_c}\right) \exp\left(-\frac{2\pi l\,\Gamma}{\hbar\omega_c}\right) J_0\left(\pi l \frac{W}{\hbar\omega_c}\right). \qquad (5)$$

This nonlinear equation cannot be solved analytically without any approximations, but it determines oscillations of the chemical potential with arbitrary parameters (it is only assumed that $\varepsilon_F \gg T, \hbar\omega_c$).

The magnetization oscillations at the constant electron density $N = \mathrm{const}$ are given by

$$M = -\left.\frac{d(\Omega+N\mu)}{dB}\right|_{N=\mathrm{const}} = -\left.\frac{\partial\Omega}{\partial B}\right|_{\mu,N=\mathrm{const}} -$$
$$-\left(\left.\frac{\partial\Omega}{\partial\mu}\right|_{N,B=\mathrm{const}} + N\right) \left.\frac{d\mu}{dB}\right|_{N=\mathrm{const}} = -\left.\frac{\partial\Omega}{\partial B}\right|_{\mu,N=\mathrm{const}}.$$

The oscillating part of the magnetization is

$$\tilde{M}(B) = -\left.\frac{\partial\tilde{\Omega}}{\partial B}\right|_{\mu,N=\mathrm{const}} =$$

$$= \frac{2g}{\pi B}\varepsilon_F \sum_{l=1}^{\infty} \frac{(-1)^{l+1}}{l} \frac{\lambda l}{\mathrm{sh}\,\lambda l} \times$$
$$\times \cos\left(2\pi l \frac{\mu_e H}{\hbar\omega_c}\right) \exp\left(-\frac{2\pi l\,\Gamma}{\hbar\omega_c}\right) \times$$
$$\times \left\{ \sin\left(2\pi l \frac{\mu(B)}{\hbar\omega_c}\right) J_0\left(\pi l \frac{W}{\hbar\omega_c}\right) + \right.$$
$$\left. + \frac{W}{2\mu} \cos\left(2\pi l \frac{\mu(B)}{\hbar\omega_c}\right) J_1\left(\pi l \frac{W}{\hbar\omega_c}\right) \right\}, \qquad (6)$$

where $\mu(B)$ is given by Eq. (5) and involves the dependence of the magnetization on the reservoir DoS. Equations (5) and (6) describe the magnetization oscillations at arbitrary parameters. The only approximation used in deriving these formulas is the Dingle law of harmonic damping. In quasi-$2D$ organic metals with the warping $W > T_D$, the Dingle law is believed to be a sufficiently good approximation.

Equations (5) and (6) are a good starting point for numerical calculations. It follows that in the limit $W/\mu \ll 1$, the oscillating parts of the magnetization and the chemical potential are related simply by

$$\tilde{M}(B) = \frac{\varepsilon_F}{B} \frac{2g}{\hbar\omega_c}(1+n_R)\tilde{\mu}(B). \qquad (7)$$

For zero warping, this was obtained in [9].

Nonlinear equation (5) for $\tilde{\mu}(B)$ can be solved analytically only in some simple approximations. We do this to illustrate the influence of the chemical potential oscillations on the temperature and the Dingle temperature dependence of the harmonic amplitudes. We thus consider zero warping, zero spin splitting and zero temperature. The sum in the right-hand side of Eq. (5) can then be calculated and we obtain

$$\frac{x}{2} = \frac{1}{(1+n_R)} \mathrm{arctg}\left(\frac{\sin(y+x)}{\cos(y+x)+e^b}\right), \qquad (8)$$

where $x \equiv 2\pi\tilde{\mu}(B)/\hbar\omega_c$, $y \equiv 2\pi\varepsilon_F/\hbar\omega_c$, and $b \equiv 2\pi\Gamma/\hbar\omega_c$.

For a very large electron reservoir $n_R = \infty$, we have $x = 0$, which implies the case of a fixed chemical potential. In this case, the magnetization is given by [13]

$$\tilde{M}(B) = \frac{2g\varepsilon_F}{\pi B} \mathrm{arctg}\left(\frac{\sin y}{e^b + \cos y}\right). \qquad (9)$$

The temperature dependence of the harmonic amplitudes is given by the LK formula

$$A_l(T) = \frac{2\pi^2 T\, l/\hbar\omega_c}{\mathrm{sh}(2\pi^2 T\, l/\hbar\omega_c)}. \qquad (10)$$

It is also possible to solve Eq. (8) analytically at $n_R = 0$ and $n_R = 1$. At zero electron reservoir $n_R = 0$, the solution of this equation is

$$\frac{x}{2} = \pi \frac{\tilde{\mu}(B)}{\hbar\omega_c} = \mathrm{arctg}\left(\frac{\sin y}{e^b - \cos y}\right).$$

It gives the oscillations of the chemical potential. The magnetization at zero electron reservoir is given by

$$\tilde{M}(B) = \frac{2g\,\varepsilon_F}{\pi B}\,\mathrm{arctg}\left(\frac{\sin y}{e^b - \cos y}\right). \tag{11}$$

It coincides with (9) after the phase shift $y \to y + \pi$ and the sign change $\tilde{M} \to -\tilde{M}$. This implies that the harmonic damping law

$$A_l \propto \frac{1}{l}\exp\left(-lb\right) \tag{12}$$

does not change and only the sign of all even harmonics is reversed. This symmetry between the cases of the fixed chemical potential $\mu = \mathrm{const}$ and the constant particle density $N = \mathrm{const}$ is a feature of the special exponential law of the harmonic damping. Any finite temperature and the electron reservoir density breaks this symmetry.

We now consider the intermediate case where $n_R = 1$. Equation (8) then becomes

$$\frac{\sin x}{\cos x} = \frac{\sin(y + x)}{\cos(y + x) + e^b}. \tag{13}$$

This gives

$$x = \arcsin\left(e^{-b}\sin y\right).$$

For the magnetization, we obtain

$$\tilde{M}(y) = \frac{g\,\varepsilon_F}{\pi B}\arcsin\left(e^{-b}\sin y\right). \tag{14}$$

To determine how the harmonic damping has changed, we must calculate the amplitudes of the first several harmonics of this expression. The amplitude of the first harmonic is

$$A_1(b) = \frac{1}{\pi}\int\limits_{-\pi}^{\pi}\arcsin\left(e^{-b}\sin y\right)\sin y\,dy,$$

and after the integration by parts, we obtain

$$A_1(b) = \frac{4}{\pi}\int\limits_{0}^{\pi/2}\frac{\cos^2 y\, e^{-b}\,dy}{\sqrt{1 - e^{-2b}\sin^2 y}}.$$

This is a superposition of two elliptic integrals,

$$A_1(b) = \frac{4}{\pi}\left[e^b E(e^{-b}) - 2\,\mathrm{sh}\,b\, K(e^{-b})\right]. \tag{15}$$

For $b \gg 1$, the deviations of $A_1(b)$ from the LK formula are small,

$$A_1(b) = e^{-b} + e^{-3b}/8 + \ldots$$

In the opposite limit $b \ll 1$, we obtain

$$A_1(b) = \frac{4}{\pi}\left\{1 - b\left(\ln\frac{4}{\sqrt{2b}} - \frac{1}{2}\right) + O(b^2)\right\}. \tag{16}$$

This is substantially different from the LK dependence $A_1(b) = \exp(-b) \approx 1 - b$. For example, the value $A_1(0)$ is $4/\pi$ times larger than the LK prediction.

A stronger deviation from LK formula (12) can be seen in the amplitudes of the next harmonics. All even harmonics vanish because expression (14) possesses the symmetries $\tilde{M}(\pi - y) = \tilde{M}(y)$ and $\tilde{M}(-y) = \tilde{M}(y)$.

The amplitude of the third harmonic can also be calculated. For $b \gg 1$ and $e^{-b} \ll 1$, we have

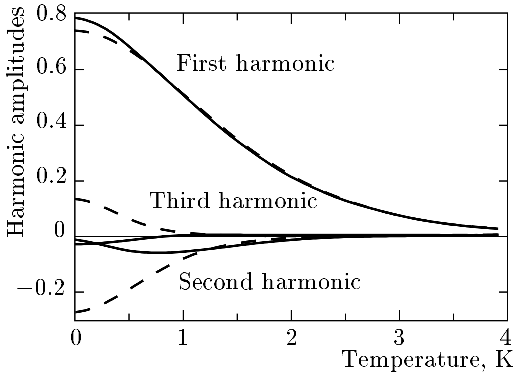
$$A_3(b) = -e^{-3b}/12 + O(e^{-5b}).$$

This result is in contrast with the cases where $n_R = 0$ or $n_R = \infty$, where we had $A_3(b) = e^{-3b}/3$. This is not surprising because in the symmetric case $n_R = 1$, the oscillations must be much smoother and more sinusoidal. Therefore, the first harmonic must increase and the higher harmonics must decrease. For $b = 0$, we have

$$A_3(0) = \frac{4}{3\pi}\int\limits_{0}^{\pi/2}\frac{\cos 3y\,\cos y\,dy}{\cos y} = -\frac{4}{9\pi}, \tag{17}$$

which is $\sim$ 2.35 times less than the LK prediction $A_3(0) = 1/3$ and has the opposite sign. In the case where $n_R = 1$, the first harmonic is therefore increased while the amplitudes of the others are strongly decreased compared to the cases of zero and infinite electron reservoir. The deviation from the LK formula reduces as the warping of the FS increases. The above analysis also shows that at low temperature and low Dingle temperature, the harmonic ratios can give a quantitative estimate of the electron reservoir density that is much more precise than just an observation about the slope of magnetization oscillations.

To include the correct temperature dependence, warping, and spin-splitting and to consider an arbitrary reservoir density, one can do numerical calculations based on solving Eq. (5) for the chemical potential and inserting this solution in formula (6) for the magnetization. This can be easily done for arbitrary parameters that are available experimentally. The temperature dependence of the first three harmonic amplitudes is given in the Figure for the following set of parameters close to the real experiments



Temperature dependence of harmonic amplitudes. The solid lines are the numerical results (for $n_R = 1$, $m^* = 2m_0$, $T_D = 0.2$ K, and $W = 1$ K; see text) and the dashed lines are the LK prediction at the same parameters. Their strong deviations are clearly seen, especially for higher harmonics

on α-(BEDT-TTF)$_2$KHg(SCN)$_4$: the reservoir density $n_R = 1$, the dHvA frequency $F = 700$ T, the effective mass $m^* = 2m_0$, the Dingle temperature $T_D = 0.2$ K, and the warping $W = 1$ K. We see a substantial deviation from the LK dependence. As $T \to 0$, the obtained amplitude of the first harmonic is about 1.1 times larger than the LK prediction. If we also let $T_D \to 0$ and $W \to 0$, their ratio becomes $4/\pi = 1.27$ in agreement with analytical result (16). The second harmonic amplitude is close to zero at $T = 0$. The amplitude of the third harmonic changes its sign at $T \approx 0.8$ K and deviates very strongly from the LK formula. It is damped much stronger than the LK predictions. At $T = 0$ and $W = 0$, it also coincides with prediction (17).

To conclude, it was shown both analytically and numerically that the oscillations of the chemical potential are essential for the temperature dependence of harmonic amplitudes of the dHvA oscillations in quasi-two-dimensional compounds. The accurate determination of the effective electron mass from the dHvA effect should take this effect into account. This can be done by a simple numerical calculation based on Eqs. (5) and (6). The oscillations of the chemical potential depend on the reservoir density of states in accordance with Eq. (5). This fact may be used for estimating the reservoir density of states in organic metals.

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