de Haas-van Alphen Effect in the Two-Dimensional and the Quasi-Two-Dimensional Systems

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We study the de Haas-van Alphen (dHvA) oscillation in two-dimensional and quasi-two-dimensional systems. We give a general formula of the dHvA oscillation in two-dimensional multi-band systems. By using this formula, the dHvA oscillation and its temperature-dependence for the two-band system are shown. By introducing the interlayer hopping \( t_z \), we examine the crossover from the two-dimension, where the oscillation of the chemical potential plays an important role in the magnetization oscillation, to the three-dimension, where the oscillation of the chemical potential can be neglected as is well known as the Lifshitz and Kosevich formula. The crossover is seen at \( 4t_z \sim 8abH/\phi_0 \), where \( a \) and \( b \) are lattice constants, \( \phi_0 \) is the flux quantum and \( 8t \) is the width of the total energy band. We also study the dHvA oscillation in quasi-two-dimensional magnetic breakdown systems. The quantum interference oscillations such as \( \beta-\alpha \) oscillation as well as the fundamental oscillations are suppressed by the interlayer hopping \( t_z \), while the \( \beta+\alpha \) oscillation gradually increases as \( t_z \) increases and it has a maximum at \( t_z/t \approx 0.025 \). This interesting dependence on the dimensionality can be observed in the quasi-two-dimensional organic conductors with uniaxial pressure.

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I. INTRODUCTION

It is well known that the magnetization \( (M) \) periodically oscillates as a function of the inverse magnetic field \( (1/H) \) with frequencies of the extremes of the cross-section area of the Fermi surface. This oscillation, known as the de Haas-van Alphen (dHvA) oscillation, is used to study the shape of the Fermi surface of the conductors and the experiments have been fitted using the Lifshitz and Kosevich (LK) formula. From the fitting of the temperature-dependence of the amplitude of the dHvA oscillation, we can obtain the cyclotron effective mass \( (m) \) for each cyclotron orbits.

Recently, a revival of interest in dHvA oscillations in two-dimensional and quasi-two-dimensional conductors arises, since it has been noticed that the LK formula cannot be applied to the two-dimensional and quasi-two-dimensional conductors. Because of the Landau quantization of the two-dimensional kinetic energy in the magnetic field, both electron number \( (N) \) and chemical potential \( (\mu) \) cannot be fixed simultaneously as the magnetic field is changed. In the actual situation, \( N \) is fixed, and \( \mu \) oscillates periodically as a function of \( 1/H \). In the three-dimensional conductors the oscillation of \( \mu \) is shown to be very small. Therefore the LK formula obtained under the condition of fixed \( \mu \) (grand canonical ensemble) is justified to be used in the system of the fixed \( N \) (canonical ensemble). However, in the two-dimensional system, the oscillation of \( \mu \) as a function of \( 1/H \) cannot be neglected. The saw-tooth waveform of the dHvA oscillation in the fixed \( N \) case is inverted from that in the fixed \( \mu \) case in the single-band case.

It has been shown that there appear the quantum interference oscillations with the frequencies such as \( f_\beta - f_\alpha \) and \( f_\beta + f_\alpha \) in the magnetization when \( N \) is fixed in magnetic breakdown systems of the tight-binding electrons. In the magnetic-breakdown systems treated semi-classically and in multi-band systems the anomalous spin-splitting effects are also shown to exist in the fixed \( N \) systems.

From numerical calculations for the single-band system, it has been shown that the temperature-dependence of the amplitude of the dHvA oscillation cannot be described by the LK formula. The conventional fitting of the LK formula may not give the cyclotron effective mass in the two-dimension.

In this paper we give a general formula for the dHvA oscillation in two-dimensional multi-band systems. From this formula, the oscillatory parts of the chemical potential and the magnetization are calculated numerically in the two-band system. Moreover, by increasing the interlayer hopping \( t_z \), we study how the dHvA oscillation in the two-dimensional system change to those in the three-
In the next section we give the general formula for the dHvA oscillation. In section IV we show the difference of dHvA oscillations in cases of the fixed $\mu$ and the fixed $N$ in the two-dimensional multi-band system. In Section III the crossover from two-dimensional to three-dimensional magnetic breakdown systems is studied. In section V the quantum interference oscillation are studied in two-dimensional and quasi-two-dimensional magnetic breakdown systems.

**II. FORMULATION**

The thermodynamic potential $\Omega$ is calculated as a function of the chemical potential $\mu$, magnetic field $H$ and temperature $T$ as

$$\Omega(\mu, H) = -T \log \left( \sum_{N,E} \exp \left( -\frac{E - \mu N}{T} \right) \right)$$

$$= -T \int d\epsilon N_0(\epsilon, H) \log \left( 1 + \exp \left( -\frac{\epsilon - \mu}{T} \right) \right),$$

(1)

where $E$ is the total energy of the system, $N$ is the number of electrons and $N_0(\epsilon, H)$ is the density of states in the presence of the magnetic field $H$. The magnetization $M$ is obtained in the grand canonical ensemble as

$$M_\mu(\mu, H) = -\frac{\partial \Omega(\mu, H)}{\partial \mu}. \quad (2)$$

As mentioned by Lifshitz and Kosevich, $\mu$ is treated as the independent variable of $H$ in the above equation. This is not the case in the actual situation.

The electron number $N$ is calculated in the grand canonical ensemble as

$$N(\mu, H) = -\frac{\partial \Omega(\mu, H)}{\partial \mu}. \quad (3)$$

In section II we show the difference of dHvA oscillations in cases of the fixed $\mu$ and the fixed $N$ in the two-dimensional multi-band system. In Section III the crossover from two-dimensional to three-dimensional magnetic breakdown systems is studied. In section V the quantum interference oscillation are studied in two-dimensional and quasi-two-dimensional magnetic breakdown systems.

**III. TWO-DIMENSIONAL ELECTRONS**

**A. multi-band system**

In this section we study the two-dimensional multi-band electrons in the magnetic field. This model can be applied to the multi-band materials such as Sr$_2$RuO$_4$ and In$_x$Ga$_{1-x}$As. The density of states in the magnetic field is given by the sum of delta functions,

$$N_0(\epsilon, H) = \sum_j \sum_{n=0}^\infty \sum_{\sigma=\pm} \rho_j(\epsilon - \epsilon_{jn\sigma}), \quad (8)$$

where $j$ is the band suffix, $\rho_j$ is the density of states for each spin in the $j$ band at $H = 0$,

$$\omega_j = \frac{eH}{m_j}, \quad (9)$$

is the cyclotron frequency with the cyclotron mass $m_j$,

$$\epsilon_{jn\sigma} = \omega_j(n + \frac{1}{2}) + g_j(\sigma \mu_B H + \Delta_j), \quad (10)$$

is the Landau level, $\Delta_j$ is the energy of the band bottom in zero magnetic field, $g_j$ is the electron $g$-factor for the $j$ band and $\mu_B = e\hbar/2m_0c_0$ is the Bohr magnetron, electron mass $m_0$ and velocity of light $c_0$. In this section, we set $h = c_0 = k_B = 1$. The electron number at $H = 0$ is given by

$$N = \sum_j \sum_{\sigma=\pm} \int_{\Delta_j}^{\mu_0} \rho_j d\epsilon = \sum_j \int_{\mu_0}^{\mu_j} \rho_j(\mu_0 - \Delta_j), \quad (11)$$

where $\mu_0$ is the chemical potential at $H = 0$.

Applying the Poisson formula in Eq. (12), thermodynamic potential is written as

$$\Omega(\mu, H) = \Omega_0(\mu, H) + \tilde{\Omega}(\mu, H), \quad (12)$$

where
\[ \Omega_0(\mu, H) = -T \int_0^\infty \frac{de}{\sigma} \sum_j \rho_j \sum_{\sigma=\pm 1/2} \rho_j \times \log \left(1 + \exp \left(\frac{\mu - \Delta_j - g_j \sigma \mu B H - \epsilon}{T}\right)\right) \] (13)

and

\[ \tilde{\Omega}(\mu, H) = \frac{1}{12} \sum_j \rho_j \omega_j^2 \]
\[ + \sum_{j=1}^\infty \sum_{r=1}^\infty \frac{\rho_j \omega_j^2}{\pi^2 r^2} \times R_{T,jr} \times R_{S,jr} \]
\[ \times \cos \left(2\pi r \left(\frac{\mu - \Delta_j}{\omega_j} + \frac{1}{2}\right)\right), \] (14)

where

\[ R_{T,jr} = \frac{2\pi r T/\omega_j}{\sinh(2\pi r T/\omega_j)} \], (15)
\[ R_{S,jr} = \cos \left(\pi r \frac{\tilde{g}_j}{2}\right), \] (16)

and

\[ \tilde{g}_j = g_j \frac{m_j}{m_0}. \] (17)

In the above we have used the identity

\[ \int_0^\infty \frac{\sin x}{1 + e^{Bx-A}} dx = 1 - \frac{\pi \cos(A/B)}{B \sinh(\pi/B)} - \sum_{n=1}^\infty \frac{(-1)^n e^{-An}}{1 + B^2 n^2}, \] (18)

where \(A = (\mu - \Delta_j - g_j \sigma \mu B H)/T, B = \omega_j/(2\pi r T)\), and we have neglected the summation term over \(n\), since we are interested in low temperature properties \((A \gg 1)\).

At low temperature \((T \ll \mu - \Delta_j)\), we get

\[ \Omega_0(\mu, H) \approx -\frac{1}{2} \sum_j \rho_j (\mu - \Delta_j - g_j \sigma \mu B H)^2. \] (19)

We obtain

\[ \frac{\partial \Omega_0(\mu, H)}{\partial H} = -\frac{\mu B}{4} \sum_j \rho_j \tilde{g}_j^2. \] (20)

From this equation, it is found that \(\partial \Omega_0(\mu, H)/\partial H\) does not make the oscillation of the magnetization as a function of \(1/H\). Thus, in grand canonical ensemble, \(\tilde{\Omega}(\mu, H)\) gives the oscillatory part of the magnetization \(\tilde{M}_\mu(\mu, H)\) as

\[ \tilde{M}_\mu(\mu, H) = -\frac{\partial \tilde{\Omega}(\mu, H)}{\partial H} \]
\[ \approx -\sum_{j=1}^\infty \sum_{r=1}^\infty \frac{2\pi^2 f_j \rho_j}{\pi^2 m_j^2} R_{T,jr} R_{S,jr} \]
\[ \times \sin \left(2\pi r \left(\frac{f_j}{H} + \frac{1}{2}\right)\right) \] (21)

where

\[ 2\pi f_j = 2\pi m_j (\mu - \Delta_j), \] (22)

is the area of the Fermi surface at \(H = 0\) in the \(j\) band. Conventionally, \(R_{T,jr}\) is known as the temperature reduction factor of the LK formula and is used to estimate the cyclotron effective mass \((m_j)\) from the \(T\)-dependence of the amplitude of the oscillation. The electron number is obtained from Eq. (3) as

\[ N(\mu, H) = -\frac{\partial \Omega_0}{\partial \mu} - \frac{\partial \Omega}{\partial \mu} \]
\[ = \sum_j 2\rho_j \left\{\mu - \Delta_j \]
\[ + \sum_{r=1}^\infty \frac{\omega_j}{\pi r} R_{T,jr} R_{S,jr} \]
\[ \times \sin \left(2\pi r \left(\frac{\mu - \Delta_j}{\omega_j} + \frac{1}{2}\right)\right)\right\}. \] (23)

Here we consider the fixed \(N\) case. As we wrote in the previous section, the above equation should be taken as the equation for \(\mu(\mu, H)\). With the use of Eq. (14), we get,

\[ \tilde{\mu}(\mu, H) = \frac{-1}{\rho} \sum_j \rho_j \omega_j \]
\[ \times \sin \left(2\pi r \left(\frac{\tilde{\mu}(\mu, H) + \mu_0 - \Delta_j}{\omega_j} + \frac{1}{2}\right)\right) \] (24)

where

\[ \rho = \sum_j \rho_j, \] (25)

and \(\tilde{\mu}(\mu, H) = \mu(\mu, H) - \mu_0\) is the oscillatory part of the chemical potential. This is a transcendental equation for \(\tilde{\mu}(\mu, H)\) and it is difficult to give \(\tilde{\mu}(\mu, H)\) by solving Eq. (24) analytically except for simple cases such as the single-band case at \(T = 0\) that we will discuss below. After we obtain \(\mu(\mu, H)\), we can calculate \(M_N(\mu, H)\) (Eq. (5)). From the second equation of Eq. (3) and Eq. (14),

\[ M_N(\mu, H) = -\left(\frac{\partial \Omega_0(\mu, H)}{\partial H} + \frac{\partial \tilde{\Omega}(\mu, H)}{\partial H}\right)_{\mu=\mu(\mu, H)}. \] (26)

It is found from Eq. (21) that \(\partial \Omega_0(\mu, H)/\partial H\) is independent of \(\mu\). Thus, the first term of of the right side of Eq. (26) does not contribute to the oscillation of \(M_N(\mu, H)\) as a function of \(1/H\). As a result, we get

\[ M_N(\mu, H) = -\left(\frac{\partial \tilde{\Omega}(\mu, H)}{\partial H}\right)_{\mu=\mu(\mu, H)} \]
\[ = \tilde{M}_\mu(\mu(\mu, H), H), \] (27)
where $\tilde{M}_N(N, H)$ is the oscillatory part of $M_N(N, H)$.

Recently, Alexandrov and Bratkovskiy\cite{30} asserted that the oscillations of the magnetization also come from $\Omega_0$ through the oscillation of $\mu$. Their result for the free energy (Eq. (13) in their paper\cite{30}) is formally correct but they did not take account of the magnetic field dependence of the $f_\alpha$, which cannot be neglected in two-dimensional systems. As a result their analysis of the dHvA oscillation for fixed $N$ system (canonical ensemble) is insufficient and their conclusions on Fourier transform intensities are incorrect.

When all of the band-bottom energies are the same ($\Delta_j = \Delta$), we can derive the simple relation between $\tilde{\mu}(N, H)$ and $\tilde{M}(N, H)$. Since $\tilde{\Omega}(\mu, H)$ rapidly oscillates as a function of $(\mu - \Delta)/H$ in this case and the envelope depends on $H$ slowly, we can approximate

$$\frac{\partial \tilde{\Omega}(\mu, H)}{\partial H} \approx -\frac{\partial \tilde{\Omega}(\mu, H)}{\partial \mu} \frac{\mu(N, H) - \Delta}{H}. \quad (28)$$

Using Eq. (4) we get

$$-\frac{\partial \tilde{\Omega}(\mu, H)}{\partial \mu} = N + \frac{\partial \Omega_0}{\partial \mu}, \quad (29)$$

and using Eqs. (15) and (19) we get

$$-\frac{\partial \tilde{\Omega}(\mu, H)}{\partial \mu} = -2\rho \tilde{\mu}(N, H). \quad (30)$$

Then Eq. (27) is rewritten as

$$\tilde{M}_N(N, H) = \frac{2}{H} \rho \mu_0 \tilde{\mu}(N, H) \left(1 - \frac{\Delta}{\mu_0} + \frac{\tilde{\mu}(N, H)}{\mu_0}\right)$$

$$\approx \frac{2\rho (\mu_0 - \Delta)}{H} \tilde{\mu}(N, H)$$

if $\Delta_j = \Delta$ for all $j$, \quad (31)

where $|\tilde{\mu}(N, H)/\mu_0| \ll 1$ is used. Thus the oscillatory part of the magnetization is proportional to the oscillatory part of the chemical potential, if all the band-bottom energies are same. The similar relation between $\tilde{M}_N(N, H)$ and $\tilde{\mu}(N, H)$ in the single-band system with electron (or hole) reservoirs has been obtained independently by Mineev and Champeaux\cite{30}, Itskovsky et al., and Grigoriev.\cite{30} However, when the band-bottom energies are not the same, Eq. (31) is not satisfied. In that case, $\tilde{M}_N(N, H)$ should be calculated from Eq. (27) with numerically solved $\tilde{\mu}(N, H)$ in Eq. (24). In order to confirm the above discussion, we show $\mu(N, H)$ and $\tilde{M}_N(N, H)$ in the cases of $\Delta_\alpha = \Delta_\beta$ and $\Delta_\alpha \not= \Delta_\beta$ in the two-band system ($j = \alpha, \beta$). We set $m_\alpha = 0.5m_0$, $m_\beta = m_0$, $g_\alpha = g_\beta = 0$, $T = 0.0001\mu_0$, $\Delta_\beta = 0$ and $\Delta_\alpha = 0, 0.34\mu_0$ and $0.68\mu_0$. By assuming the parabolic band, $\rho_\beta = m_\beta/2\pi$. The sum over $r$ in Eqs. (24) and (27) is taken up to 30. We have checked that the results are the same within the numerical errors when we take the sum over $r$ up to 50. It can be clearly seen from Figs. 1 and 2 that $\tilde{\mu}(N, H)$ and $\tilde{M}_N(N, H)$ oscillate as the same function of $H$ when $\Delta_\alpha = \Delta_\beta$. When $\Delta_\alpha$ is not equal to $\Delta_\beta$, $\tilde{\mu}(N, H)$ is different from $\tilde{M}_N(N, H)$, as seen in Figs. 2 and 3.

![FIG. 1. $\tilde{\mu}(N, H)$ (dotted line) and $\tilde{M}_N(N, H)$ (solid line) as a function of $\mu_0/\omega_\beta$ when $\Delta_\alpha = \Delta_\beta = 0$.](image1)

![FIG. 2. The same as Fig. 1 when $\Delta_\alpha = 0.34\mu_0$ and $\Delta_\beta = 0$.](image2)

![FIG. 3. The same as Fig. 1 when $\Delta_\alpha = 0.68\mu_0$ and $\Delta_\beta = 0$.](image3)
In Fig. 4 we show $\tilde{M}_N(N, H)$, $\tilde{M}_\mu(\mu, H)$ and their Fourier transform intensities (FTIs), where we set $m_\alpha = 0.5m_0$, $m_\beta = m_0$, $\Delta_\alpha = 0.68\mu_0$, $\Delta_\beta = 0$, $g_\alpha = g_\beta = 0$ and $T = 0.0001\mu_0$. With these parameters the frequencies are $f_\alpha = m_\alpha(\mu_0 - \Delta_\alpha)/e = 0.16m_0\mu_0/e$ and $f_\beta = m_0\mu_0/e$. These parameters are the same as in the previous studies except for the temperature; the ground state energy is calculated directly by filling Landau levels at $T = 0$ in the previous studies. It is seen from Fig. 4 that the FTI has large peaks at the combination frequencies $f_\beta + f_\alpha$, $f_\beta + 2f_\alpha$, $2f_\beta + 2f_\alpha$ and $f_\beta - f_\alpha$ besides at the fundamental frequencies $f_\beta$ and $f_\alpha$ which are in agreement with previous results at $T = 0$. These combination oscillations are known as the quantum interference oscillations. The quantum interference oscillations are caused by the oscillation of $\mu$. If $\mu$ is fixed, as seen in dotted line in Fig. 4 the FTI of $\tilde{M}_\mu(\mu, H)$ has peaks at the fundamental frequencies ($f_\beta$ and $f_\alpha$) and higher harmonics ($2f_\beta$ and $2f_\alpha$) and it has no peaks at the combination frequencies.

![Fig. 4. $\tilde{M}_N(N, H)$ and $\tilde{M}_\mu(\mu, H)$ (lower figures) as a function of $\mu_0/\omega_\beta$ and their Fourier transform intensities (upper figures), where the frequency has the unit of $m_0\mu_0/e$. The Fourier transform is performed in the region of $255 \leq \mu_0/\omega_\beta \leq 273$.](image)

We show the $T$-dependence of the FTIs of $\tilde{M}_\mu(\mu, H)$ and $\tilde{M}_N(N, H)$ in Fig. 4 where the parameters are the same as those in Fig. 4. In order to take in the sharpness of the oscillation at the very low temperature, we set the upper limit of $r$ in Eqs. (23) and (24) as 75 below $T/\mu_0 = 0.000003$. The $T$-dependence of the FTI is given by $R_{T,jr}^2$ (Eq. (13)) when $\mu$ is fixed. When $N$ is fixed, the temperature dependence of FTIs are no longer given by $R_{T,jr}^2$. Although the temperature dependence of the FTI at $f_\alpha$ is well fitted by $R_{T,jr}^2$, the temperature dependences of the FTIs at $f_\beta$, $f_\beta \pm f_\alpha$ are not fitted by the temperature reduction factors, $R_{T,jr}^2$. Therefore, we should be careful to determine the cyclotron mass from the temperature dependence of the FTIs in two-dimensional systems.

![Fig. 5. $T$-dependence of the FTIs of the magnetization at $f_\alpha$, $f_\beta$, $f_\beta - f_\alpha$ and $f_\beta + f_\alpha$ in the fixed $N$ case (solid lines) and FTIs at $f_\alpha$ and $f_\beta$ in the fixed $\mu$ case (dotted lines).](image)

**B. special case: single-band system at $T = 0$**

In the single-band ($\rho_j = \rho$, $\omega_j = \omega$, $m_j = m$ and $\Delta_j = \Delta$) and no-spin-splitting case ($g_j = 0$) at $T = 0$, we can obtain $\tilde{M}_N(N, H)$ analytically. By using the identity

$$[x] = x - \frac{1}{2} \sum_{r=1}^{\infty} \frac{1}{\pi r} \sin (2\pi r x),$$

where $[x]$ is the largest integer satisfying $[x] \leq x$, Eq. (23) at $T = 0$ is written as

$$N(\mu, H) = 2\rho \omega \left[ \frac{\mu - \Delta}{\omega} + \frac{1}{2} \right],$$

When we take this equation as an equation giving $\mu(N, H)$, we get

$$\mu(N, H) - \Delta = \left( \frac{N}{2\rho \omega} + \frac{1}{2} \right) \omega.$$  

This equation means that $\mu$ is pinned at the Landau level and it jumps from one Landau level to the other when $N/(2\rho \omega)$ is integer. Since $(\mu(N, H) - \Delta)/\omega + 1/2$ is integer, we obtain from Eq. (14) that $\Omega(\mu(N, H), H)$ does not oscillate as a function of $H$ in the single-band case at
\[ T = 0 \text{ when } N \text{ is fixed. By using the identity (Eq. (32)) in Eq. (34), we get} \]
\[ \mu(N, H) = \frac{N}{2\rho} + \Delta + \sum_{r=1}^{\infty} \frac{\omega}{\pi r} \sin \left( \frac{2\pi r f}{\bar{H}} \right), \quad (35) \]

where \( f = Nm/(2pe) \).

It should be noticed that if Eq. (24) is considered as an equation giving \( \mu(N, H) \) in the fixed \( N \) case, this equation is not satisfied when Eq. (34) is inserted: If we insert Eq. (34) in Eq. (24), we get the right hand side as \( 2\rho(\mu(N, H) - \Delta) \) for the single-band case, because \( (\mu(N, H) - \Delta)/\omega + 1/2 \) is integer in Eq. (34). Although \( \mu(N, H) \) oscillates as a function of \( H \), the left hand side of Eq. (23) should be independent of \( H \) when \( N \) is fixed. Therefore, we cannot insert Eq. (34) obtained at \( T = 0 \) into Eq. (23). The reason for this paradoxical result is the following. The Landau level with the energy \( \mu(N, H) \) is partially filled at \( T = 0 \). This information is lost in Eq. (24). In the same way we cannot get \( \tilde{\Delta} \) in the fixed \( N \) case. Therefore, we cannot insert Eq. (34) obtained at \( T = 0 \) into Eq. (23). The relation Eq. (31) is always satisfied in the single-band case even at \( T = 0 \). From Eqs. (21) and (33), we get

\[ \tilde{\Delta}(N, H) = \frac{eN}{m} \sum_{r=1}^{\infty} \frac{1}{\pi r} \sin \left( \frac{2\pi r f}{\bar{H}} \right). \quad (36) \]

We see from Eq. (21) and Eq. (33) that the oscillation of the magnetization for the fixed \( N \) has a phase-shifted and sign-changed saw-tooth shape comparing with that obtained in the fixed \( \mu \) in the two-dimensional single-band spinless case at \( T = 0 \).

**IV. QUASI-TWO-DIMENSIONAL TIGHT-BINDING ELECTRONS**

In this section, we study the crossover from two-dimension to three-dimension in the quasi-two-dimensional electrons at \( T = 0 \). If we do not take into account the periodicity of the system, the system has the closed Fermi surface even for the very large effective mass in the \( z \)-direction. Therefore, in order to study the crossover from two-dimension (with cylindrical Fermi surface) to three-dimension we have to study the periodic system.

We take the tight-binding electrons in the simple orthogonald lattice with lattice constants \( a, b \) and \( c \) and the hopping \( t_x, t_y \) and \( t_z \). For simplicity we neglect the spin (i.e. \( g = 0 \)) and set \( t_x = t_y = t \). The Hamiltonian in the absence of the magnetic field is given by

\[ \hat{H}_0 = \sum_{\mathbf{k}} \hat{c}^\dagger(\mathbf{k}) \mathcal{E}(\mathbf{k}) \hat{c}(\mathbf{k}), \quad (37) \]

where

\[ \mathcal{E}(\mathbf{k}) = -2t(\cos ak_x + \cos bk_y) - 2t_z \cos ck_z. \quad (38) \]

The magnetic field is introduced by the Peierls substitution,

\[ \mathbf{k} \rightarrow \mathbf{k} + e\mathbf{A}, \quad (39) \]

where \( \mathbf{A} \) is a vector potential. In this paper the magnetic field is taken to be along the \( z \)-direction and the Landau gauge is used,

\[ \mathbf{A} = (Hy, 0, 0). \quad (40) \]

Then the Hamiltonian is written as

\[ \hat{H} = -t \sum_{\mathbf{k}} \{ \exp(ikx) \hat{c}^\dagger(k_x, k_y - \delta, k_z) \hat{c}(\mathbf{k}) + h.c. \} \]

\[ + \sum_{\mathbf{k}} (-2t \cos(bk_y) - 2t_z \cos k_z) \hat{c}^\dagger(\mathbf{k}) \hat{c}(\mathbf{k}). \quad (41) \]

where

\[ \delta = \frac{eaH}{\hbar c_0} = \frac{\phi}{\phi_0} \frac{2\pi}{b}, \quad (42) \]

\( \phi = abH \) is the flux passing through a unit cell, and \( \phi_0 = 2\pi\hbar c_0/e \) is the unit flux quantum. For the numerical calculation we take the number of the flux quantum per plaquette,

\[ h = \frac{\phi}{\phi_0} \quad (43) \]

to be a rational number. We use \( h \) instead of \( H \) hereafter. If we take \( a = b \approx 10 \text{ [Å]} \) (the typical values for quasi-two-dimensional organic conductors), \( h \approx 1/40 \) corresponds to \( H = 100 \text{ [T]} \). We obtain the eigenvalues \( (\epsilon_i(h)) \) by diagonalizing \( \hat{H} \) numerically. Then we calculate the Helmholtz free energy \( F(N, h) \) at \( T = 0 \), which is just the total energy, as

\[ F(N, h) = \frac{1}{N_s} \sum_{i=1}^{N} \epsilon_i(h), \quad (44) \]

where \( N_s \) is the site number. We fix the electron number to be 1/3 filled, i.e. \( N/N_s = 1/3 \). The Fermi surface at \( h = 0 \) and \( t_z = 0 \) is shown in Fig. 4. The area of the closed Fermi surface is

\[ f = \frac{1}{3} f_{\text{BZ}}, \]

where \( f_{\text{BZ}} = 4\pi^2/ab \). The magnetization is obtained from the numerical differential

\[ M_N(N, h) = -\frac{\delta F(N, h)}{\delta h}. \quad (45) \]

The chemical potential is given by

\[ \mu(N, h) = \epsilon_N(h). \quad (46) \]
FIG. 6. Fermi surface of the two-dimensional tight-binding electrons with filling factor $1/3$.

In the tight-binding model, even in the two-dimensional case ($t_z = 0$), the density of states is not the sum of the delta functions but it consists of mini-bands with the finite width (Harper broadening). However, the width is very small especially in the case of $t_a = t_b$ as shown in Fig. 7. On the other hand, the mini-gap, which means the spacing between the Landau levels, is of the order of $8t$, as indicated by dotted lines in Fig. 7. (The total band width is of the order of $8t$, which consists of $q$ delta-function-like mini-bands when $h = p/q$ with co-prime integers $p$ and $q$. However, $p$ mini-bands are very close each other, making one super-mini-band. Therefore, the mini-gap is roughly of the order of $8t/(q/p) = 8t$. For example, consider $h = 1/40$ and $h = 3/118$. These two cases should have similar density of states as can be seen in Fig. 7. Indeed, they have the similar mini-band-gap, $8t \times (1/40) \approx 8t \times (3/118)$, if we neglect the mini-mini-gap inside the super-mini-band which consists of three mini-bands in the case of $h = 3/118$.)

When $t_z$ becomes finite, the density of states becomes broader as shown in Fig. 8. The width of each mini-band becomes the order of $4t_z$. If the width of the mini-band is the same order of the mini-gap at $t_z = 0$, that is, $4t_z \approx 8t$, the density of states has no energy-gap as in the three-dimensional systems in the magnetic field. We expect the crossover from two-dimension to three-dimension occurs at $h \approx t_z/2t$. In fact, we can see this crossover from the density of states for $t_z/t = 0.04$ and $1/h = 40$, as shown in Fig. 8. The actual dimensional crossover in the quasi-two-dimensional organic conductors may occur at $H \approx 20 \text{[T]}$. As, in these organic conductors, $8t$ is of the order of 1 [eV] and $t_z$ is of the order of 10 [K $\mu$], the crossover field is $h \approx t_z/2t \approx 1/200$, that is, $H \approx 20 \text{[T]}$. 

FIG. 7. Density of states for the tight-binding electrons in the magnetic field in the two-dimensional system ($t_z = 0$). The arrows show the chemical potential for $1/3$-filled band.

FIG. 8. Density of states for the tight-binding electrons in the magnetic field in the quasi-two-dimensional system ($t_z/t = 0.04$). The arrows show the chemical potential for $1/3$-filled band.
When the system is two-dimensional ($t_z = 0$), the chemical potential is pinned in the narrow region of the non-zero density of states as in the case of the two-dimensional free electrons. In this case, the chemical potential and the magnetization oscillate periodically as a function of $1/h$, as shown by the dotted lines in Fig. [4]. The frequency of the oscillation of $\mu(N, h)$ and $M_N(N, h)$ is $1/3$, which is in agreement with the closed Fermi surface area in the unit of the Brillouin zone, $f/f_{BZ}$.

In the quasi-two-dimensional system, the amplitudes of the oscillations of $\mu(N, h)$ and $M_N(N, h)$ become smaller as $t_z$ increases as seen in Fig. 2. In particular, the oscillation of $\mu(N, h)$ is very small when the density of states becomes gapless ($h = 1/40$ and $t_z/t = 0.04$ as shown in Fig. 8) where $4t_z$ is nearly equal to $8th$, i.e., $4t_z/(8th) = t_z/(2th) = 0.8$.

Fig. 9. $\mu$ (upper figure) and $M_N(N, h)$ (lower figure) as a function of $1/h$. Arrows indicate the magnetic field for which the density of states are plotted in Figs. 7 and 8.

We compare the magnetizations in the fixed $N$ and fixed $\mu$ cases in Fig. 4. The magnetization with fixed $\mu$ at $T = 0$ is obtained from the numerical differential of the thermodynamic potential $\Omega(\mu, h)$ as

$$M_\mu(\mu, h) = -\frac{\delta\Omega(\mu, h)}{\delta h}, \quad \text{(47)}$$

where

$$\Omega(\mu, h) = \frac{1}{N_h} \sum_{\epsilon_i \leq \mu} (\epsilon_i(h) - \mu). \quad \text{(48)}$$

The chemical potential is fixed to be $\mu/2t = -0.414$ to satisfy $N/N_h = 1/3$ at $h = 0$. In Fig. 4, the magnetization is plotted as a function of $1/h$ for both cases of fixed $\mu$ and fixed $N$. We calculate $M_N(N, h)$ and $M_\mu(\mu, h)$ for two-dimensional case ($t_z = 0$) and quasi-two-dimensional case ($t_z/t = 0.04$). As expected, for the two-dimensional case ($t_z = 0$), two curves have inverted and shifted saw-tooth shape each other, which can be seen from the upper figures in Fig. 4.

In the quasi-two-dimensional case, the difference between $M_N(N, h)$ and $M_\mu(\mu, h)$ can be seen at $h > 1/24$ and it becomes small at smaller field as clearly seen in Fig. 10. This is consistent with the behavior of $\mu(N, h)$ shown in Fig. 3, i.e. the amplitude of the oscillation of $\mu(N, h)$ becomes small when $t_z/(2t) \gtrsim h$.

![FIG. 10. $M_N(N, h)$ and $M_\mu(\mu, h)$ as a function of $1/h$. Arrows indicate the magnetic field for which the density of states are plotted in Figs. 7 and 8.](image)

V. QUANTUM INTERFERENCE OSCILLATION IN QUASI-TWO-DIMENSIONAL ELECTRONS

In this section we study the dHvA oscillation in the quasi-two-dimensional systems which has the small cylindrical Fermi surface and the quasi-one-dimensional Fermi surface, as shown in Fig. 11. This Fermi surface is realized in $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ and GaAs/AlAs heterointerface. The small cylindrical Fermi surface is separated from the quasi-one-dimensional Fermi surface on the first Brillouin zone edge (dotted lines in Fig. 11) by the periodic potential. In the semiclassical picture, electrons are expected to execute a large closed orbital motion by tunneling the first Brillouin zone gap at high fields. Then, the oscillation due to the large closed orbit appears in the dHvA oscillation. This is known as the magnetic breakdown phenomena. In $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ and $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$, the magnetic breakdown is observed.
Fig. 11. Fermi surface of the two-dimensional tight-binding electrons in the presence of periodic potential with filling factor $7/18$. The area of the electron pocket centered at $(0, \pm \pi/2)$ is $f_{\alpha}/f_{BZ} \approx 1/18$ in the unit of the area of the extended Brillouin zone $(2\pi)^2$. The orbit enabled by the magnetic breakdown has the area $f_{\beta}/f_{BZ} \approx 7/18$.

In order to obtain this Fermi surface we add the periodic potential

$$\hat{V} = V \sum_{r=(a_{i}, b_{i}, c_{i})} (-1)^{r} \hat{c}^{\dagger}(r) \hat{c}(r)$$

$$= V \sum_{k} \{ \hat{c}^{\dagger}(k + \mathbf{g}) \hat{c}(k) + \text{h.c.} \},$$

where

$$\mathbf{g} = (0, \frac{\pi}{b}, 0),$$

Fig. 12. $M_{N}(N, h)$ and $M_{\mu}(\mu, h)$ (lower figure) as a function of $1/h$ and their Fourier transform intensities (upper figure) at $t_{z} = 0$. The Fourier transform is performed in the region of $12 \leq 1/h \leq 62$. The peaks of the FTI occur at $f_{\alpha}$, $f_{\beta}$, $f_{\beta} - f_{\alpha}$ and $f_{\beta} + f_{\alpha}$ besides smaller peaks at $2f_{\alpha}$, $f_{\beta} \pm 2f_{\alpha}$ etc.

We calculate the magnetization and its FTIs numerically as in the previous section. In Fig. 12, we show the results in the two-dimensional case. It can be seen from the lower figure in Fig. 12 that the amplitude of the oscillation with the frequency $f_{\beta}$ becomes large due to magnetic breakdown as $1/h$ becomes small. In the magnetic breakdown system, the oscillation with $f_{\beta} + f_{\alpha}$ is expected to exist but the oscillation with $f_{\beta} - f_{\alpha}$ is forbidden in the semiclassical picture. This semiclassical picture is consistent with our quantum calculation as seen in the dotted line in the upper figure in Fig. 12, as known previously.

If the two bands are not connected with magnetic breakdown, combination frequencies ($f_{\beta} \pm f_{\alpha}$ etc.) do not appear in $M_{\mu}(\mu, H)$, as can be seen from dotted lines in Fig. 14. On the other hand, in the two-band system, both frequencies $f_{\beta} \pm f_{\alpha}$ are seen in $M_{N}(N, H)$ (Fig. 13), where the FTI at $f_{\beta} + f_{\alpha}$ is larger than that at $f_{\beta} - f_{\alpha}$. In the magnetic breakdown system, however, the FTI at $f_{\beta} + f_{\alpha}$ in $M_{N}(N, H)$ is very small as seen in the solid line in the upper figure in Fig. 12. This behavior has also been known previously.

In the previous papers we have shown that the FTI at $f_{\beta} + f_{\alpha}$ in $M_{N}(N, H)$ in the magnetic breakdown system is anomalously enhanced by spin-splitting effect. We have interpreted the anomalous spin-enhancement of the FTI at $f_{\beta} + f_{\alpha}$ in $M_{N}(N, H)$ in the magnetic breakdown system as follows. From the results of two-band and magnetic breakdown systems, it is found that the oscillation with $f_{\beta} + f_{\alpha}$ is caused by both effects of the magnetic breakdown and the chemical potential oscillation, as can be seen in the fixed $\mu$ case in Fig. 12 and in the fixed
case in Fig. 4, respectively. If the electron spin is neglected in magnetic breakdown systems in the fixed N case, these two origins of the $f_{\beta} + f_{\alpha}$ oscillation almost cancel each other resulting in the small FTI at $f_{\beta} + f_{\alpha}$ in $M_N(N, H)$. The spin-splitting may reduce the chemical potential oscillation. As a result, the cancellation becomes imperfect and the FTI at $f_{\beta} + f_{\alpha}$ in $M_N(N, H)$ in the magnetic breakdown system is enhanced due to the spin-splitting.

The similar effect is expected in the case of quasi-two-dimensional system. As $t_z$ is increased, the chemical potential oscillation is suppressed, resulting in the enhancement of the FTI at $f_{\beta} + f_{\alpha}$. This is indeed the case as we show in Figs. 13 and 14. In Fig. 13 we plot $M_N(N, h)$ and its FTIs for $t_z/t=0, 0.01, 0.02$ and 0.03. As $t_z$ increases, the FTIs at $f_{\alpha}$, $f_{\beta}$ and $f_{\beta} - f_{\alpha}$ decrease as in the single-band case studied in the previous section. The FTI at $f_{\beta} + f_{\alpha}$, however, depends very weakly on $t_z$; it gradually increases as $t_z$ increases, and it has a maximum around $t_z/t \approx 0.025$ as shown by the arrow in Fig. 14.

VI. CONCLUSION

We find that the dHvA oscillation in two-dimensional multi-band systems is given by Eq. (27) with the chemical potential obtained by solving Eq. (24). It is the general formula, i.e., it can be applied in the multi-band systems at finite temperatures including the electron spin. In two-dimensional systems, the cyclotron effective mass is generally not obtained by fitting the temperature dependence with the reduction factor $R_{T,jr}$. It should be obtained from the fitting by Eq. (27) with Eq. (24).

We also study the crossover of the dHvA oscillation from two-dimensional systems to three-dimensional systems. In the simple case of the single Fermi surface we show that the oscillation of the magnetization calculated in the canonical ensemble (fixed $N$) approaches smoothly to that calculated in the grand canonical ensemble (fixed $\mu$). In the magnetic breakdown system we find that the Fourier transform intensity at $f_{\beta} + f_{\alpha}$ gradually increases as a three-dimensionality increases and it has a maximum at $t_z/t \approx 0.025$. The $t_z$ dependence can be observed experimentally by applying the uniaxial stress in the quasi-two-dimensional organic conductors, since the uniaxial stress along the $z$-axis will increase $t_z$.

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