Quantum Interference Oscillation in DHvA Effect in the Two- and Three-dimensional Systems

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We study the behavior of the DHvA oscillation from the two-dimensional to the three-dimensional by changing the interlayer hopping $t_z$. We show that the oscillation of the chemical potential is gradually suppressed as $t_z$ increases. It results from the broadening of the width of the Landau level. In the quasi-two-dimensional magnetic breakdown system such as $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, the quantum interference oscillation such as $\beta$-$\alpha$ oscillation is suppressed, while the amplitude of the $\beta$-$\alpha$ oscillation has a maximum as a function of $t_z$. This interesting dependence on the dimensionality can be observed in quasi-two-dimensional metals under the uniaxial pressure.

KEYWORDS: DHvA oscillation, magnetic breakdown, quasi-two-dimensional system, $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, Sr$_2$RuO$_4$, chemical potential oscillation

§1. Introduction

In the metal, the magnetization $(M)$ periodically oscillates as a function of the inverse of the magnetic field $(H)$, which is known as the de Haas-van Alphen (DHvA) oscillation.\(^1\) The experiment of the DHvA oscillation has been conventionally examined by the Lifshitz-Kosevich (LK) formula.\(^2\) When we neglect the impurity scattering in the spinless case and at $T = 0$, the LK formula is written as

$$M_{LK} \propto -\sum_{i=1}^{\infty} \sum_{p=1}^{\infty} a_i(p, H) \sin \left( 2\pi p \frac{f_i}{H} + \frac{1}{2} \right),$$

where $f_i = A_i(cH/2\pi e)$ and $A_i$ is the area of the extremal closed Fermi surface for each orbit $(i)$. As the three-dimensionality and the value of $p$ increase, $a_i(p, H)$ becomes small. This LK formula is derived from the oscillatory part of $M(\mu, H) = -\partial \Omega(\mu, H)/\partial H$, where $\Omega(\mu, H)$ is the thermodynamic potential obtained under the condition of the fixed chemical potential $(\mu)$. However, indeed, $M$ should be calculated as $M(N, H) = -\partial F(N, H)/\partial H$ by using of the Helmholtz free energy, $F(N, H)$, obtained under the condition of the fixed electron number $(N)$. In the three-dimensional system, as the width of the Landau level broadens (see Fig. 1(a)), the oscillation of $\mu$ as a function of $H$ becomes very small, so that the LK formula treating $\mu$ independent of $H$ is justified.

However, in the two-dimensional system, the LK formula is not appropriate because $\mu$ oscillates as a function of $H$ due to the pinning of $\mu$ at the very sharp Landau level (see Fig. 1(b)). Thus, the calculation under the condition of the fixed $N$ is needed. In fact, in the one-band model, Shoenberg\(^3\) has showed that the oscillatory part of $M(N, H)$ for the fixed $N$ is given by

$$M_{N}^{2D} \propto \sum_{p=1}^{\infty} \frac{1}{p} \sin \left( 2\pi p \frac{f_i}{H} \right),$$

where the spin and the impurity scattering are neglected at $T = 0$. This $M_{N}^{2D}$ is not in agreement with the LK formula $(M_{LK})$ in which the three-dimensionality is reduced.

In the two-dimensional magnetic breakdown system,\(^3-8\) it has been shown that the $\beta$-$\alpha$ oscillation (so-called the quantum interference oscillation) exists in $M(N, H)$ under the condition of the fixed $N$.\(^8,9\) Experimentally, the $\beta$-$\alpha$ oscillation was confirmed in the quasi-two-dimensional organic conductors\(^21\) such as $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$\(^22,23\) and $\alpha$-(BEDT-TTF)$_2$KHz(SCN)$_4$\(^24\) with the magnetic breakdown Fermi surface. The conventional FS theory based on the LK formula could not explain the $\beta$-$\alpha$ oscillation.

Next, we consider the quasi-two-dimensional system with the weak $k_z$-dispersion given as $-t_z \cos k_z$.\(^25\) We introduce $2t_z/\hbar \omega$ as a parameter which determines the dimensionality under the magnetic field applied to $k_z$-axis. Then, two limits can be understood as follows:

(1) When $2t_z/\hbar \omega \gtrsim 1.0$, the oscillation of $\mu$ is very small

(2) When $2t_z/\hbar \omega \ll 1.0$, the oscillation of $\mu$ is very small

Fig. 1. Schematic density of the states in the magnetic field
since the density of states becomes nearly the continuum. We regard this case as the effective three-dimensional system, where the LK formula is valid.

(2) For $2t_z/h\omega \approx 0$, we can neglect the three-dimensionality. Thus, the theory for two-dimensional system\textsuperscript{1,6-20} is appropriate.

However, the theoretical and experimental studies of the dHvA oscillation from the two- to the effective three-dimensional system (that is, in $0 \leq 2t_z/h\omega \approx 1$) have never been done.\textsuperscript{26,27} This study is very important to understand the experiments of the dHvA oscillation of the actual quasi-two-dimensional materials such as κ-(BEDT-TTF)$_2$Cu(NCS)$_2$\textsuperscript{28,29} and Sr$_2$RuO$_4$.

In this paper, by the numerical calculations under the condition that $N$ is fixed, we show $\mu$ and $M$ in the dimensionless crossover region, where the oscillation of $\mu$ is gradually suppressed and $M$ is smoothly changed (see Fig. 2). In the magnetic breakdown system, we find the anomalous enhancement of the amplitude of the $\beta+\alpha$ oscillation as a function of $t_z$ (see Fig. 5).

\section*{§2. Formulation}

We use a tight binding model. In the $x$-$y$ plane, the nearest neighbor transfer integrals ($t_x$ and $t_y$) with the lattice spacing ($a$ and $b$) and an on-site potential ($V$) are considered. For $V = 0$, a closed Fermi surface exists, and we call it as $\beta$ orbit and define its area as $f_\beta$ in the unit of $4\pi^2/ab$ (see Fig. 3 (a)). For small $V$, there exist a small closed Fermi surface (called as $\alpha$ orbit) and the quasi-one-dimensional open Fermi surface, where the large $\beta$ orbit is possible due to the magnetic breakdown (see Fig. 3 (b)). From this model, we can study both the one-band system ($V = 0$) and the magnetic breakdown system ($V \neq 0$) in the $z$-axis. We consider the weak interlayer hopping ($t_z$) with the lattice spacing ($c$). For simplicity, we take $t_x = t_y = t$ and ignore the spin of the electrons to avoid additional complications. The magnetic field is applied in the $z$-direction.

![Fig. 2. $\mu(N, h)$ (upper figure) and $M(N, h)$ (lower figure) as a function of $1/h$ in the one-band model.](image)

![Fig. 3. (a) The Fermi surface at $k_z = 0$ for the one-band model ($V = 0$). (b) for magnetic breakdown model ($V \neq 0$). The Hamiltonian is written as\textsuperscript{25}]

$\hat{\mathcal{H}}(h) = \hat{\mathcal{K}}(h) + \hat{\mathcal{V}}$.\textsuperscript{(2.1)}

Without magnetic field in $k$-space representation,

$\hat{\mathcal{K}}(0) = \sum_k \hat{\mathcal{C}}^\dagger(k)\mathcal{E}(k)\hat{\mathcal{C}}(k)$,\textsuperscript{(2.2)}

$\hat{\mathcal{V}} = V \sum_k \left\{ \hat{\mathcal{C}}^\dagger(k, k_x + g)\hat{\mathcal{C}}(k) + \text{h.c.} \right\}$,\textsuperscript{(2.3)}

$\mathcal{E}(k) = -t(\cos ak_x + \cos bk_y) - t_z \cos ck_z$,\textsuperscript{(2.4)}

where $g = \pi/b$.

The electron filling $(N/N_s)$ is set to about $1/3$ and $7/18$ for $V = 0$ and $V \neq 0$, where $N_s$ is the total site number. In this case, $f_\beta \approx 1/3$ for $V = 0$, and $f_\alpha \approx 1/18$, $f_\beta \approx 7/18$ and $f_{\beta + \alpha} = f_\beta + f_\alpha$ for $V \neq 0$.

We introduce the magnetic field by the Peierls substitution $k \rightarrow k + eA/h$ in $\mathcal{E}(k)$, taking the Landau gauge $A=(Hy, 0, 0)$. The resultant kinetic energy operator is

$\hat{\mathcal{K}}(h) = -\frac{i}{2} \sum_k \left\{ \exp(ikz)\hat{\mathcal{C}}^\dagger(k_0, k_x - \delta)\hat{\mathcal{C}}(k) + \text{h.c.} \right\}$

$+ \sum_k \left\{ -t \cos(bky) - t_z \cos ck_z \right\}$\textsuperscript{(2.5)}

$\hat{\mathcal{C}}^\dagger(k)\hat{\mathcal{C}}(k)$, where $\delta = \frac{eA}{hc} = \frac{2\pi}{\phi_0} = \frac{2\pi}{b}$, $\phi = abH$ is the flux passing through a unit cell, $\phi_0 = 2\pi/e$ is the unit flux quantum, and $h = \phi/\phi_0$ is the number of the flux quantum per unit cell. Since the total band width is about $4t_z$, $h\omega \approx 4th = 4h$, and $2t_z/h\omega \approx t_z/2h$. When $t_z/t = 0.05$, $2t_z/h\omega \approx 1$ at $h \approx 1/40$. We describe the magnetic field as $h$, hereafter. In this study, $h$ is changed from $1/60$ to $1/8$, which corresponds to $20 \approx n \approx 3$, where $n$ is the number of the highest occupied Landau level.

From the eigenvalue ($E_i(h)$) obtained by diagonalizing $\mathcal{H}(h)$, the chemical potential is given by

$\mu(N, h) = E_B(h)$\textsuperscript{(2.6)}

The free energy, $F(N, h)$, at $T = 0$ is given by

$F(N, h) = \frac{1}{N_s} \sum_{i=1}^{N} E_i(h)$\textsuperscript{(2.7)}

The magnetization is obtained from

$M(N, h) = -\partial F(N, h)/\partial h$\textsuperscript{(2.8)}
§3. Results and Discussions

3.1 One-band Model

In Fig. 2, we show $\mu(N, h)$ and $M(N, h)$ under the condition of the fixed $N$ as a function of $1/h$ by changing $t_z$ continuously. The period of the oscillation of $\mu(N, h)$ and $M(N, h)$ is 1/3, which is consistent with the period of $f_3$. The effective three-dimensionality due to the larger $t_z$ and/or the larger $h$ results in the smooth damping of the oscillation of $\mu(N, h)$ and the smooth decreasing of the amplitude of the oscillation of $M(N, h)$. When the three-dimensionality becomes large (see the case of $t_z/t = 0.05$ in Fig. 2), the oscillation of $\mu(N, h)$ is very small at the low field ($1/h \gtrsim 30$), i.e. $2t_z/h \gtrsim 1.0$.

3.2 Magnetic Breakdown Model

In the magnetic breakdown system ($V/t = 0.1$), $M(N, h)$ and its Fourier transform amplitudes (FTAs) are shown in Fig. 4. From the overall behavior of $M(N, h)$, we can see that the amplitudes of the oscillations of $\alpha$ ($\beta$) are smaller (larger) as $h$ increases, which is due to the magnetic breakdown.

In the FTAs of $M(N, h)$ there exit the $\beta$-$\alpha$ oscillation and a small peak of $\beta+\alpha$ oscillation in addition to the large peaks of $\alpha$ and $\beta$ oscillations at $t_z = 0$. In Fig. 5, we show the $t_z$-dependence of these FTAs. As $t_z$ increases, the $\alpha$, $\beta$, and $\beta+\alpha$ oscillations are strongly suppressed. The three-dimensionality plays a role of the decreasing amplitude of the oscillation. Nevertheless, the $\beta+\alpha$ oscillation is enhanced clearly and has a maximum at $t_z/t \approx 0.025$. The damping of the oscillation of $\mu$ is closely related to this maximum of the $\beta+\alpha$ oscillation.

§4. Conclusion

We have studied the dHvA oscillation in the quasi-two-dimensional system. In the intermediate region ($0 < 2t_z/h \omega < 1.0$), the effect of the smooth damping of the oscillation of $\mu$ appears in the dHvA oscillation. This interesting physical picture has never been shown until now. One can see an anomalous maximum of the amplitude of the $\beta+\alpha$ oscillation in the quasi-two-dimensional magnetic breakdown system. It will be observed in $\kappa$-

Fig. 4. $M(N, h)$ (lower figure) and its FTA (upper figure). The region of the Fourier transform is $12 \leq 1/h \leq 62$.

Fig. 5. Main FTAs of $M(N, h)$ as a function of $t_z/t$. An arrow indicates a maximum.

(BEDT-TTF)$_2$Cu(NCS)$_2$ by the experiments under the uniaxial stress.\(^{30}\)

3) The magnetic breakdown phenomena is known as follows: As the higher field enables an electron to tunnel the first Brillouin zone gap and to move along larger closed orbit, the oscillation due to this larger orbit appears. On the basis of Pippard's network model,\(^{4}\) Falicov and Stacho\'vski\(a\) (FS)\(^{5}\) derived a semiclassical formula in which the tunneling process is taken account in the LK formula.
25) $t_z$ is much smaller than $t$, where $t$ is the transfer integral in the $k_x$-$k_y$ plane. The width of the Landau level is given by $2t_z$. The Landau level spacing is $h\omega$, where $\omega = eH/c\mu$, $\mu$ is speed of light and the cyclotron effective mass, respectively.
26) Grigoriev and Vagner have shown the envelope of $M(N, H)$ in the quasi-two-dimensional one-band system.\(^{27}\) They do not show overall $M(N, H)$ and $\mu(N, H)$ as a function of $H$.
28) In quasi-two-dimensional organic conductors with $m/m_e \sim 2$, $t_z$ is of the order of a few $K$,\(^{28}\) where $m_e$ is free electron mass. Thus, $0 \leq 2t_z/h\omega \leq 1$ corresponds to $H \leq 20$ [T].