Enhancement of de Haas-van Alphen Oscillation due to Spin in the Magnetic Breakdown System

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The effects of the Zeeman term on the de Haas-van Alphen oscillation is studied in the magnetic breakdown system. We find that the amplitude of the oscillation with the frequencies of \( f_\beta + f_\alpha \) and \( f_\beta + 2f_\alpha \) are enhanced by the Zeeman term, while they are expected to be reduced in the semiclassical theory. A possible interpretation of the experiments in organic conductors is discussed.

KEYWORDS: magnetic breakdown, quantum interference oscillation, spin, quantum magnetic oscillation, quasi-two-dimensional organic conductors

§1. Introduction

The magnetic breakdown is observed in the Shubnikov-de Haas oscillation (for the transport quantity) and the de Haas-van Alphen oscillation (for the thermodynamics quantity). These oscillations are so-called the magnetic quantum oscillation, which is attributed to the Landau quantization in the closed orbit in quasi-two-dimensional organic conductors such as \( \kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2 \) and \( \alpha-(\text{BEDT-TTF})_2\text{MgHg(SCN)}_4 \) \((M=\text{K, Rb, Tl and NH}_4)\), many experiments for the magnetic breakdown are done, since those materials have the suitable Fermi surface (Fig. 1) for the study of the magnetic breakdown. In this case, the electron orbital motion due to magnetic fields \( (H) \) applied perpendicular to \( k_x-k_y \) plane is confined to the small closed orbit named as the \( \alpha \) orbit and a pair of open orbits at low fields as shown in Fig. 1. Then, this closed orbital motion leads to the Landau quantization, so that the magnetization and magnetoresistance oscillate by the frequency \( (f_\alpha) \) of the area of the \( \alpha \) orbit as a function of \( 1/H \). The high field enables electrons to tunnel the Brillouin zone gap and to move along the larger closed orbit named as the \( \beta \) orbit. Thus, the oscillation frequency \( (f_\beta) \) of the area of the \( \beta \) orbit gradually appears as \( H \) increases. This is the magnetic breakdown, which has been explained in a semiclassical theory by Falicov and Stachoivak based on Pippard’s network model. In Falicov and Stachoivak theory, the oscillations with various frequencies \( (f_\alpha, f_\beta, f_\beta-2f_\alpha, f_\beta+2f_\alpha, f_\beta+f_\alpha, \text{etc.}) \) are allowed, since the closed orbital motion is possible. However, the orbit named as the \( \beta-\alpha \) orbit in Fig. 1 is prohibited, because the \( \beta-\alpha \) orbit is not allowed to execute the cyclic motion in the semiclassical picture. The oscillation corresponding to the area of prohibited orbits such as the \( \beta-\alpha \) orbit is called the quantum interference oscillation. Shiba and Fukuyama have shown the existence of the quantum interference oscillation in the transport in the case of the magnetic breakdown. It is now well-known as the Stark quantum interference oscillation. On the other hand, it was believed that the quantum interference oscillation in the thermodynamics quantity dose not exist.

However, by using the simple tight-binding model one of the authors first found the quantum interference oscillation, that is, the \( \beta-\alpha \) oscillation in the magnetization \( M(H,n) \), whose frequency \( (f_{\beta-\alpha} = f_\beta - f_\alpha) \) corresponds to the area of the \( \beta-\alpha \) orbit in Fig. 1 by calculating the free energy, \( E(H,n) \), full-quantum-mechanically at the fixed total electron number, \( n \).

Recently, the existence of the \( \beta-\alpha \) oscillation in the magnetization was experimentally observed in \( \kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2 \) \((\text{M=K, Rb, Tl and NH}_4)\) and \( \alpha-(\text{BEDT-TTF})_2\text{MgHg(SCN)}_4 \). From other aspects of theory, the existence of the \( \beta-\alpha \) oscillation in \( M(H,n) \) was confirmed. Nakanda and Alexandrov and Bratkovsky account for the \( \beta-\alpha \) oscillation in the magnetization \( M(H,n) \) by using two independent energy band model neglecting the magnetic breakdown. Harrison et. al calculated by the density of states derived from Pippard
network model and found the $\beta$-\alpha oscillation. In addition they reveal that there is no $\beta$-\alpha oscillation in the magnetization $M(H, \mu)$ when the chemical potential, $\mu$, is fixed. In other words, they clarify that the quantum interference oscillation such as the $\beta$-\alpha oscillation is caused by the chemical potential oscillation. In fact in our model, in the case of the fixed chemical potential, the $\beta$-\alpha oscillation in $M(H, \mu)$ did not appear. Moreover, Sandu et al. and Han et al. found the $\beta$-\alpha oscillation in $M(H, n)$ from the full-quantum-mechanical calculation by use of more realistic tight-binding model based on the band calculation.

In our spinless tight-binding model, the energy spectrum becomes so-called Hofstadter’s butterfly diagram in the presence of magnetic field. Various oscillations with $f_\alpha, f_\beta, f_{2\beta}, f_{\beta-\alpha}$, etc., in $M(H, n)$ are due to the chemical potential oscillating. In that calculation the oscillation with $f_{\beta+\alpha}$ ($\beta+\alpha$ oscillation) is much smaller than $\beta-\alpha$ oscillation, although the $\beta+\alpha$ oscillation is obtained to be larger than the $\beta-\alpha$ oscillation in the independent band model. The origin of the suppression of the $\beta+\alpha$ oscillation in the magnetic breakdown system is not clear yet, but since the $\beta+\alpha$ oscillation is large in the case of the fixed chemical potential, a cancellation between the effects of the magnetic breakdown and the chemical potential oscillation will cause the suppression of the $\beta+\alpha$ oscillation. If we take account of the spin, the energy spectrum will be divided into two energy bands. Even if the total number of electrons is fixed, the electron number for each spin is not fixed. As a result we may expect a richer effect than the reduction factor in the semiclassical theory by taking account of the effect of spin in the system with the magnetic breakdown. However, it has never been studied that how the spin affects the “prohibited” quantum interference oscillation ($\beta$-\alpha oscillation) and quantum magnetic oscillations ($\alpha, \beta, \beta+\alpha, 2\beta$ oscillations, etc.). Therefore, we need to investigate the effect of the spin on the magnetic breakdown and quantum interference oscillation.

In the thermodynamics quantity such as the magnetization, $\beta+\alpha$ oscillation are observed in addition to $\alpha, 2\alpha, \beta-\alpha, \beta$ and $2\beta$ oscillations in the quasi-two-dimensional organic conductors. However, the peak at $f_{\beta-\alpha}$ reported by Meyer et al. is very small, while that peak is large in the experiment by Uji et al. Both de Haas van Alphen experiments measured at the same material, $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, are done at the same field region ($22 \text{ T} \sim 30 \text{ T}$) and the same temperature ($\sim 0.4 \text{ K}$). The difference in the amplitudes of $\beta-\alpha$ oscillation may be attributable to the effect of spin in a tilted magnetic field.

In this paper, we calculate $M(H, n)$ from the energy, $E$, at the canonical ensemble by adding the Zeeman term to the previous spinless tight-binding model and try to analyze the effect of the spin on the magnetic breakdown and the quantum interference oscillation.

\section{Formulation}

Let us study the two-dimensional tight-binding model. The lattice spacings in $x$- and $y$-axes are $a$ and $b'$, respectively. The Hamiltonian at $H = 0$ is

$$\mathcal{H} = \mathcal{K} + \mathcal{V}. \quad (1)$$

$$\mathcal{K} = \sum_{i,j,\sigma} t_{ij} C_{i,\sigma}^\dagger C_{j,\sigma} = \sum_{\mathbf{k},\sigma} C_{\mathbf{k},\sigma}^\dagger \epsilon(\mathbf{k}, \sigma) C_{\mathbf{k},\sigma}. \quad (2)$$

$$\mathcal{V} = V \sum_{i,\sigma} \cos \mathbf{Q} \cdot \mathbf{r}_i C_{i,\sigma}^\dagger C_{i,\sigma}. \quad (3)$$

In the above $\sigma = \uparrow, \downarrow$, $C_{\mathbf{k},\sigma}$ is the creation operator of $\sigma$ spin electron at $i$ site, $\epsilon(\mathbf{k}, \sigma) = -2t_a \cos a \mathbf{k}_x - 2t_b \cos b' \mathbf{k}_y$ and $\mathbf{Q} = (0, \pi/b')$ is the lattice potential vector, where the Brillouin zone is $-\pi/a \leq k_x < \pi/a$, $-\pi/b' \leq k_y < \pi/b'$. In the momentum space, eq. (3) is written as

$$\hat{\mathcal{V}} = \frac{V}{2} \sum_{k_x, k_y', m, \sigma} \{C_{k_x, k_y'}^\dagger (m + 1) \frac{\pi}{b'}, \sigma) \times C_{k_x, k_y'} (m \frac{\pi}{b'}, \sigma) + \text{h.c.} \}, \quad (4)$$

where summation in $k_y'$ and $m$ are done in $-\pi/2b' \leq k_y' < \pi/2b'$ and $m = 0, 1$, respectively. By $\hat{\mathcal{V}}$, the Brillouin zone is halved so that the first Brillouin zone becomes $-\pi/a \leq k_x < \pi/a$, $-\pi/b \leq k_y < \pi/b$, where we choose $b = 2b'$. In this paper, we take parameters as $2/3$-filling, $t_b/t_a = 1.0$ and $V/t_a = 0.09$. Then, the Fermi surface is obtained as shown in Fig. 1, which has a characteristic topology in quasi-two-dimensional organic conductors. From this Fermi surface we can see that the possible cyclotron orbits are the closed $\alpha$ orbit and the breakdown $\beta$ orbit.

When the magnetic field is applied parallel to $z$ axis, we take the Landau gauge $\mathbf{A} = (0, Hx, 0)$, and we write $\mathcal{K}$ by Peierls substitution as

$$\hat{\mathcal{H}} = -\sum_{k'_x, k', n, m, \sigma} \left\{ -2t_a \cos (a k'_x + n \delta) - \frac{1}{2} \mu B H \right\}$$

$$C_{k'_x + n \delta, k_y' + m \frac{\pi}{b'}, \sigma} C_{k'_x, n \delta, k_y' + m \frac{\pi}{b'}, \sigma}$$

$$+ \{ -t_b \exp[i b k'_y + i m \pi] C_{k'_x + n \delta, k_y' + m \frac{\pi}{b'}, \sigma}$$

$$C_{k'_x + (n + 1) \delta, k_y' + m \frac{\pi}{b'}, \sigma} + \text{h.c.} \}, \quad (5)$$

where $\delta = e a H / \hbar c = (\phi / \phi_0) (2 \pi / a), \phi = ab' H$ is the flux passing through a unit cell and $\phi_0 = 2 \pi \hbar c / e$ is a unit flux, and $\phi = \phi / \phi_0$ is the number of the flux quantum per unit cell. We represent magnetic fields by $h$, henceforth. When $h$ is a rational number, namely, $h = \phi / \phi_0 = p / q$ with $p$ and $q$ being mutually prime integers, the matrix size of $\hat{\mathcal{H}}$ is $q \times q$. In eq. (5), $0 \leq k'_x < \pi/p$ and $0 \leq n < q$. As a result, the matrix size of $\hat{\mathcal{H}} + \mathcal{V}$ becomes $2q \times 2q$. In $\hat{\mathcal{H}} + \mu B$ is the Bohr magneton and $g \simeq 2$. If the Zeeman term in $\hat{\mathcal{H}} + \mathcal{V}$ is excluded, this model becomes the previous spinless model.

Under the condition of the fixed total electron number, $n$, the ground state energy per site is calculated by

$$E(h, n) = \frac{1}{N_s} \sum_{j=1}^{n} \epsilon_j, \quad (6)$$

where $N_s$ is the total site number and $\epsilon_j$ is one electron eigenvalue, and index $j$ includes the spin index. The
magnetization is given by $M(h, n) = -\partial E(h, n)/\partial h$.

The magnetic field cannot be changed continuously in this formulation and the experimentally accessible field ($H \sim 10$ T) is difficult to study due to the large value of $q$. For example, as $a = 12.7\AA$ and $b = 8.4\AA$ for (BEDTTTF)$_2$X $h = p/q$ is about 1/400 when $H \sim 10$ T. We calculate at higher field ($H \sim 100$ T, $h \sim 1/40$).

In order to see the effect of the Zeeman term clearly, we define $\bar{g}$ as $\bar{g}h \equiv g\mu BH/t_a$. For example, in quasi-two-dimensional organic conductors with the transfer integral, $t_a = 250k_B$ and the lattice spacings mentioned above $(a = 12.7\AA$ and $b = 8.4\AA$) we get $\bar{g} \approx g \approx 2.0$.

Next, from the Fourier transform of $M(h, n)$ for $\bar{g} \neq 0$ (Fig. 3(b)~(d)), we can see that the peak at $f_{\beta+\alpha}$ becomes large at $\bar{g} = 0.2$, and this peak is the same order of magnitude as at $f_{\beta}$, whereas the peak at $f_{\beta-\alpha}$ is very small at $\bar{g} = 0.5$. When $\bar{g} = 0.8$, peaks at $f_{\alpha}$ and $f_{\beta+2\alpha}$ become very small, whereas peaks at $f_{2\alpha}$ and $f_{\beta+2\alpha}$ become large, where the $\beta - 2\alpha$ oscillation is the quantum interference oscillation like $\beta - \alpha$ oscillation. In order to understand the $\bar{g}$-dependence of these peaks, we show amplitudes of these peaks at $f_{\alpha}$, $f_{2\alpha}$, $f_{\beta}$, $f_{\beta+\alpha}$ and $f_{\beta+2\alpha}$ as a function of $\bar{g}$ from $\bar{g} = 0$ to 1.0 in Fig. 4.

In semiclassical theory, the effect of the spin is represented by the reduction factor $R_s = \cos(\gamma p\pi m/m_0)$, where $p$ is for $p$th harmonics of the each frequency in $M(h, n)$ and $m$ ($m_0$) are cyclotron effective mass (free electron mass). For example, when we choose $m/m_0 = 2$, $p = 1$, $t_a = 250k_B$, $a = 12.7\AA$ and $b = 8.4\AA$, the Fourier transform amplitude (FTA) of $M(h, n)$ is reduced by the factor $|R_s|^2 = |\cos(\gamma \bar{g})|^2$. We show $|R_s|^2$ in Fig. 4, where we take $\gamma = 0.001$. From Fig. 4, the $\bar{g}$-dependences of the FTAs of $f_{\alpha}$, $f_{2\alpha}$, $f_{\beta}$, $f_{\beta-\alpha}$ and $f_{\beta-2\alpha}$ decrease as $\bar{g}$ increases, and all of these except $f_{\alpha}$ increase.

§3. Result and Discussion

We show $M(h, n)$ at $\bar{g} = 0, 0.2, 0.5$ and 0.8 in Fig. 2, and the Fourier transform of $M(h, n)$ in Fig. 3.

Fig. 2. Magnetization as a function of the inverse field.

First, we see Figs. 2(a) and 3(a) when $\bar{g} = 0$. In Fig. 3(a), these peaks ($f_{\alpha} \approx 0.08$, $f_{2\alpha} \approx 0.16$, $f_{4\alpha} \approx 0.32$, $f_{3\beta} \approx 0.67$, $f_{3\beta} \approx 1.33$ and $f_{3\beta-\alpha} \approx 0.59$) correspond to the area of $\alpha$, $2\alpha$, $4\alpha$, $\beta$, $2\beta$ and $\beta-\alpha$ orbits (the ratios of area of $\alpha$ and $\beta$ orbits to the first Brillouin zone area are 0.08 and 0.67, respectively). The oscillations of the long period ($f_{\alpha}$) and that of the short period ($f_{\beta}$) in $M(h, n)$ are seen in Fig. 2(a). As $h$ increases, the amplitude of the oscillation with $f_{\beta}$ becomes gradually larger. This behavior is due to the magnetic breakdown. Although the $\beta-\alpha$ oscillation in addition to $\alpha$, $\beta$ and $2\beta$ oscillations exists, a peak for $\beta+\alpha$ oscillation is very small. This property has been shown previously by one of the authors and Harrison et al. also have obtained the similar result.

Fig. 3. The Fourier transform amplitude (FTA) of $M(h, n)$. The range of the Fourier transform is $10 \leq h^{-1} \leq 60$. 
upon further increasing $\tilde{g}$. These FTAs are in agreement with $|R_2|^2$ qualitatively. It may be due to the different values of the effective masses and $p$ that the position of the minimum of the FTAs of these frequencies are different. However, note that the $\tilde{g}$-dependences of the FTAs of $f_{\beta+\alpha}$ and $f_{\beta+2\alpha}$ are quite different from $|R_2|^2$; these amplitudes first increase as $\tilde{g}$ increases and decrease as $\tilde{g}$ increases further.

The anomalous $\tilde{g}$-dependences can be understood as follows. In the spinless case ($\tilde{g} = 0$), amplitudes of $\beta + \alpha$ and $\beta + 2\alpha$ oscillations are suppressed by the cancelation of the magnetic breakdown and the chemical potential oscillation. By finite $\tilde{g}$ the chemical potential oscillation is strongly suppressed. The suppression of the chemical potential oscillation is consistent with a strong reduction of $\beta - \alpha$ and $\beta - 2\alpha$ oscillations in small $\tilde{g}$ as seen in Fig. 4. On the other hand, the magnetic breakdown is not affected by $\tilde{g}$. As a result, $\beta + \alpha$ and $\beta + 2\alpha$ oscillations becomes large as $\tilde{g}$ increases due to an incomplete cancelation of the chemical potential oscillation and the magnetic breakdown.

When the field is tilted from the $k_z$ axis to the $k_x$-$k_y$ plane by $\theta$, the component of the magnetic field for the Fermi surface along the $k_z$ axis is $H \cos \theta$. That for the Zeeman term is not changed by tilting of $H$. Therefore, the effect of magnetic fields for the Zeeman term is enhanced by tilting the field.

As seen in Fig. 4 the amplitude of $\beta - \alpha$ oscillation depends on $\tilde{g}$, i.e., the tilting angle $\theta$. Therefore, we expect that the different results between Meyer et al. and Uji et al. are due to a direction of magnetic fields in both experiments. In fact, the frequency of $\beta$ oscillation in both experiments are not the same. The relative magnitudes of the amplitude of each frequencies ($f_{\alpha}$, $f_{\beta}$, $f_{\beta-\alpha}$, $f_{\beta+\alpha}$, etc.) are varied by changing $\tilde{g}$. Particularly, the oscillations with $f_{\beta+\alpha}$ and $f_{\beta+2\alpha}$ have the anomalous $\tilde{g}$-dependences. Thus, we should consider the system of the magnetic breakdown with the effect of the spin, which has not been taken into account in the previous studies.

§4. Conclusion

From the full-quantum mechanical calculation by using the simple tight-binding model including an electron spin, we analyze the effect of the spin on the quantum interference oscillation such as $\beta - \alpha$ and $\beta - 2\alpha$ oscillations and the quantum magnetic oscillation such as $\alpha$, $2\alpha$, $\beta$, $\beta + \alpha$ and $\beta + 2\alpha$ oscillations. It is obtained that the relative magnitudes of all oscillations ($\alpha$, $\beta$, $\beta + \alpha$, $\beta - \alpha$, etc.) are changed by the Zeeman term. In particular, the amplitudes of $\beta + \alpha$ and $\beta + 2\alpha$ oscillations have anomalous $\tilde{g}$-dependences.

We expect that the $\tilde{g}$-dependences of the amplitudes of these oscillations in our result will be observed in the experiment of tilting magnetic field.

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