Spin Splitting in de Haas-van Alphen Oscillation in Two-Dimensional Two-Band Systems

Keita KISHIGI, Yasumasa HASEGAWA and Mitake MIYAZAKI

Faculty of Science, Himeji Institute of Technology, Akou-gun, Hyogo 678-1297, Japan
(Received November 15, 2021)

We study the effects of the Zeeman term on the de Haas van Alphen (dHvA) oscillation in two-dimensional two-band systems. We found that the Fourier transform amplitudes of the oscillations are not described by the spin reduction factor in the Lifshitz-Kosevich formalism in two-dimensional systems. The anomalous dependence on the effective g-factor can be observed by tilting-angle dependence of the dHvA oscillation in quasi-two-dimensional organic conductors and Sr₂RuO₄.

KEYWORDS: dHvA Oscillation, Sr₂RuO₄, Quasi-Two-Dimensional Organic Conductors, Zeeman effect, Magnetic Breakdown

The magnetization and magnetoresistance oscillate as a function of the inverse of the magnetic field \((H)\) with the period proportional to the extreme area \((f)\) of the closed orbit on the Fermi surface. These phenomena are known as the Shubnikov-de Haas (SdH) oscillation and the de Haas-van Alphen (dHvA) oscillation. The dHvA oscillation can be described in the semiclassical theory known as the Shubnikov-de Haas (SdH) oscillation and the Fermi surface. These phenomena are caused by the phase shift in the oscillation for up and down spins. When the Zeeman splitting becomes a half of the Fermi surface, the tilt of the magnetic field.

The magnetization for the fixed electron number is given by

\[
M^s(\mu, H) = -\sum_{p=1}^{\infty} R_p^s \frac{1}{p} \sin p\left(\frac{f}{H} - \pi\right),
\]

where \(R_p^s\) is the reduction factor due to electron spin, \(g \approx 2\) is the g-factor, \(m\) is the cyclotron effective mass and \(m_0\) is the free electron mass. The spin factor is caused by the phase shift in the oscillation for up and down spins. When the Zeeman splitting becomes a half of the Landau level spacing, the amplitude of the oscillation of the fundamental frequency \((p = 1)\) becomes zero. Eq. (1) is called the LK formula. The experiments are performed in the multi energy-band system with the fixed electron number \((N)\), where the chemical potential varies as a function of the magnetic field. However, the oscillation of the chemical potential is very small when the Fermi surface has a three-dimensional shape. Then we can apply eq. (1) or the superposition of eq. (1) with some frequencies even for the system with fixed electron number.

On the other hand, if the system is two-dimensional or the interlayer coupling is much smaller than the Landau level spacings (this condition is satisfied in the quasi-two-dimensional system in the strong magnetic field), the oscillation of the chemical potential plays important roll. The magnetization for the fixed electron number is given by

\[
M^s(N, H) = \sum_{p=1}^{\infty} R_p^N \frac{1}{p} \sin p\frac{f}{H},
\]

in the two-dimensional single-band system. The spin reduction factor in this case is

\[
R_p^N = \cos\left(\pi \frac{pgm/m_0}{2}\right),
\]

where \(gm/2m_0\) is an integer part of \(gm/2m_0\). The spin reduction factor, \(R_p^N\), is the periodic function of \(gm/2m_0\) but is not the cosine function.

If the magnetic field is tilted by \(\theta\) from the z-axis, the spin reduction factor is given by replacing \(g\) with \(g/\cos\theta\) in eqs. (2) and (4). The reduction factor for the \(p\)th harmonics, \(R_p^N\), is zero when \(\cos\theta = (pgm/m_0)/(2n + 1)\) with integer \(n\). When \(\cos\theta = (gm/m_0)/(2n + 1)\), \(R_p^N\) for odd \(p\) is zero. The tilt angle, \(\theta\), when \(R_p^N\) becomes zero is called spin splitting zero condition, which can be measured by tilting the magnetic field.

When two energy bands exist in the two-dimensional system, the magnetization is not described by the superposition of the magnetization for the single band (eq. (3)) if the electron number is fixed. This is called a breakdown model. One of the authors, Sandu et al. 10 Han et al. 11 and Fortin et al. 12 treat the magnetic breakdown model as shown in Fig. 1(a), where the \(\beta\) orbit is obtained by the tunneling through the zone gap. Nakand and Alexandrov and Bratkovsky 13 have calculated the dHvA oscillation by using the independent two-band model, in which there exist two closed orbits \((\alpha\) and \(\beta\) orbits) as shown in Fig. 1 (b). From these calculations, it has been shown that there exist frequencies in the magnetization \((M(N, H))\), which are forbidden in the semiclassical theory. One of these forbidden frequencies corresponds to the area of the \(\beta\)-\(\alpha\) orbit. On the other hand, the forbidden oscillations do not appear in \(M(\mu, H)\) when \(\mu\) is fixed. These forbidden oscillations are caused by the chemical
potential oscillation.

The existence of the $\beta-\alpha$ oscillation in the magnetization has been observed experimentally in quasi-two-dimensional materials with two or three energy bands, $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$, Sr$_2$RuO$_4$, $\alpha$-(BEDT-TTF)$_2$ (SCN)$_4$ and $\kappa$-(BEDT-TTF)$_2$ (SCN)$_2$Cl$_2$. In this case neither chemical potential oscillation is neglected. In order to study how the spin affects the “forbidden” $\beta-\alpha$ oscillation and quantum magnetic oscillations ($\alpha$, $\beta$, $\beta+\alpha$, $2\alpha$ oscillations, etc.) in the two-dimensional magnetic breakdown systems, we have studied $M(N, H)$ in the tight-binding model quantum-mechanically. This model can be applied to the quasi-two-dimensional organic conductors. We found that the Fourier transform amplitudes (FTAs) of the $\beta+\alpha$ oscillation is enhanced due to the Zeeman effect, which is quite different from the spin reduction factors, eq. (2) or eq. (4). However, as far as we know, the effect of the Zeeman term in the two-dimensional two-band model has never been discussed systematically.

In this letter, we calculate $M(N, H)$ at $T = 0$ for the two-dimensional two-band model, where the magnetic breakdown is neglected. In this case neither chemical potential nor electron number for each band and each spin are fixed. Our purpose is to make clear how the oscillation in $M(N, H)$ is modulated by an electron spin.

We study the model, whose energy at $H = 0$ is given by

$$E_i(\mathbf{k}) = \frac{\hbar^2 k_x^2}{2m_\alpha} + \frac{\hbar^2 k_y^2}{2m_\beta} + \epsilon_i^h,$$

where $\epsilon_i^h$ is the bottom energy of the energy band and $i = \alpha$ and $\beta$ are the band index. The density of states for spin $\rho_i$ and the cyclotron effective mass $m_i$ are

$$\rho_i = C m_i, m_i = \sqrt{m_\alpha^2 m_\beta^2},$$

where $C = S/(2\pi \hbar^2)$ and $S$ is the real space area of the two-dimensional system.

When the magnetic field ($H_0$) is applied with the tilt-angle $\theta$, the $z$-component of the magnetic field is $H = H_0 \cos \theta$. The energy spectrum is quantized as

$$\epsilon_i(H, n, \sigma) = \hbar \omega_i(n + 1/2 + \sigma m_i \bar{g}_i) + \epsilon_i^h,$$

where $n$ is Landau index, $\omega_i = e H/m_i c$, $\sigma = \pm 1/2$, and $\bar{g}_i = g_i/2 \cos \theta$. We take $g_\alpha = g_\beta$ and $\bar{g}_i = \bar{g}$, although $g_\alpha$ and $g_\beta$ are not necessarily the same due to the spin-orbit coupling. We set $e = \hbar = c = 1$ and $C = 1$.

When the electron number ($N$) is fixed, we have to calculate the Helmholtz free energy $F(N, H)$, which is given by

$$F(N, H) = \sum_i \rho_i \hbar \omega_i \sum_{n, \sigma} \epsilon_i(H, n, \sigma),$$

where the summations are performed for the filled and partially filled energy levels. The electron number, $N$, is decided by the chemical potential, $\mu(0)$, at $H = 0$. The magnetization is obtained by

$$M(N, H) = -\partial F(N, H)/\partial H.$$
eV. In these parameters, the area of the \( \alpha \) and \( \beta \) orbits corresponds to \( f_\alpha = 160 \) and \( f_\beta = 1000 \).

We calculate the FTAs of \( M(N, H) \), where there exist many combination frequencies, \( \beta \pm \alpha, \beta \pm 2\alpha, 2\beta \pm \alpha, 2\beta \pm 2\alpha \), in addition to \( \alpha, \beta, 2\alpha \) and \( 2\beta \), which are shown in Fig. 3.

We show the FTAs of \( M(N, H) \) as a function of the magnetic breakdown model (Fig. 4 in ref. [19]). The suppression of the \( \beta + \alpha \) oscillation in the magnetic breakdown model might be understood as follows. The suppression of the \( \beta \) oscillation is caused by the cancellation due to the \( \pi \) phase difference between the oscillations for the up spin and the down spin in the semiclassical picture. This does not result in the cancellation of the \( \beta + \alpha \) oscillation in the magnetic breakdown systems because the \( \beta + \alpha \) oscillation corresponds to the larger orbit caused by the tunneling and the phase difference between the up and down spins in the \( \beta + \alpha \) orbit may not be \( \pi \). In the two-band systems without the magnetic breakdown, the \( \beta + \alpha \) oscillation is not caused by the larger orbit but the chemical potential oscillation. As a result, the \( \beta + \alpha \) oscillation is suppressed in the two-band systems when \( \alpha \) or \( \beta \) oscillation is suppressed.

The FTA of the \( 2\beta + 2\alpha \) oscillation is constant as a function of \( \tilde{g} \), which can be seen in Figs. 5 and 7.

By tilting the magnetic field the suppression of the peak of the \( \beta \pm \alpha \) oscillations and the constant peak of the \( 2\beta + 2\alpha \) oscillation as a function of \( \tilde{g} \) may be observed in \( \text{Sr}_2\text{RuO}_4 \).

The Yamaji effect[20] should be also considered when the magnetic field is tilted if the Fermi surface has weak three-dimensionality. Nakane[20] shows that the \( \beta \pm \alpha \) oscillations become large due to the Yamaji effect when \( \alpha \) and \( \beta \) oscillations are enhanced in the spinless model. In quasi two-dimensional materials, the interplay between
the spin effect studied in this paper and the Yamaji effect should be considered.

Ohmichi et al. [21] have measured the FTAs of each oscillations in the magnetoresistance in Sr$_2$RuO$_4$, where there is no suppression of the peak of the $\beta \pm \alpha$ oscillations due to the spin although the enhancement of each oscillations due to Yamaji effect is seen. Our theory for the magnetization cannot be compared with their magnetoresistance experiment [22], because the Stark quantum interference oscillations due to Yamaji effect is seen. Our theory for the spin although the enhancement of each oscillations is observed, the Zeeman effect for the two-dimensional two-band model, however, the spin splitting zero condition for the single-band (eq. (4)), whereas in $M(\mu, H)$ is given by eq.(2). Even in the two-band model, however, the spin splitting zero condition for fundamental frequencies ($\alpha$ and $\beta$ oscillations) in $M(N, H)$ is given by eq. (4). We expect that the $\bar{g}$-dependences of the amplitudes of these oscillations may be observed in the experiment of tilting magnetic field in two-dimensional multi-band system such as Sr$_2$RuO$_4$.

In conclusion, we study the dHvA oscillation including the Zeeman effect for the two-dimensional two-band model, where the magnetic breakdown is neglected. We find the anomalous $\bar{g}$-dependences of the FTAs of $\beta+\alpha$, $\beta-\alpha$ and $2\beta+2\alpha$ oscillations in $M(N, H)$, where $\bar{g} = (g(m))/(2m_0 \cos \theta)$. The $\beta \pm \alpha$ oscillations are suppressed when the $\alpha$ or $\beta$ oscillation disappears, and the FTA of the $2\beta+2\alpha$ oscillation is constant as a function of $\bar{g}$. The effect of the spin on $M(N, H)$ cannot be described by the spin reduction factor for the single-band (eq. (4)), whereas that in $M(\mu, H)$ is given by eq.(2). Even in the two-band model, however, the spin splitting zero condition for fundamental frequencies ($\alpha$ and $\beta$ oscillations) in $M(N, H)$ is given by eq. (4). We expect that the $\bar{g}$-dependences of the amplitudes of these oscillations may be observed in the experiment of tilting magnetic field in two-dimensional multi-band system such as Sr$_2$RuO$_4$.

We would like to thank M. Nakano for valuable discussions. One of the authors (K. K.) was partially supported by Grant-in-Aid for JSPS Fellows from the Ministry of Education, Science, Sports and Culture. K. K. was financially supported by the Research Fellowships of the Japan Society for the Promotion of Science for Young Scientists.

[1] I. M. Lifshitz and A. M. Kosevich: Sov. Phys. JETP 2 (1956) 636.
[2] D. Shoenberg: Magnetic oscillations in metals (Cambridge University Press: Cambridge, 1984).
[3] K. Kishigi, Y. Hasegawa and M. Miyazaki: J. Phys. Soc. Jpn. 68 (1999) 1817.
[4] M. Nakano: preprint.
[5] K. Machida, K. Kishigi and Y. Hori: Phys. Rev. B 51 (1995) 8946.
[6] K. Kishigi, M. Nakano, K. Machida, and Y. Hori: J. Phys. Soc. Jpn. 64 (1995) 3043.
[7] K. Kishigi: J. Phys. Soc. Jpn. 66 (1997) 910.
[8] N. Harrison, J. Caulfield, J. Singleton, P. H. P. Reinders, F. Herlach, W. Hayes, M. Kurmoo and P. Day: J. Phys.: Condens. Matter 8 (1996) 5415.
[9] P. S. Sandu, J. H. Kim, and J. S. Brooks: Phys Rev. B 56 (1997) 11566.
[10] So-Y. Han, J. H. Kim, and J. S. Brooks: ICSM98 proceedings.
[11] J. Y. Fortin and T. Ziman: Phys. Rev. Lett. 80 (1998) 3117.
[12] M. Nakano: J. Phys. Soc. Jpn. 66 (1997) 19.
[13] A. S. Alexandrov and A. M. Bratkovsky: Phys. Lett. A 234 (1997) 53.
[14] Quasi-two-dimensional organic conductors ($\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ and $\alpha$-(BEDT-TTF)$_2$K$_2$Hg(SCN)$_4$) have the two energy bands as shown in Fig. 1(a). In quasi-two-dimensional material, Sr$_2$RuO$_4$, three closed orbits($\alpha$, $\beta$ and $\gamma$) exist. The contribution of the magnetic breakdown for the Fermi surface of Sr$_2$RuO$_4$ is much smaller than that of the quasi-two-dimensional organic conductors.
[15] F. A. Meyer, E. Steep, W. Biberacher, P. Christ, A. Leif, A. G. M. Jansen, W. Joss, P. Wyder and K. Andres: Europhys. Lett. 32 (1995) 681.
[16] S. Uji, M. Chaparala, S. Hill, P. S. Sandhu, J. Qualls, L. Seger, and J. S. Brooks: Synth. Met. 85, (1997) 1573.
[17] M. M. Honold, N. Harrison, M. S. Nam, J. Singleton, C. H. Mielke, M. Kurmoo, and P. Day: Phys. Rev. B 58 (1998) 7560.
[18] R. Settai, Y. Yoshida, A. Mukai and Y. Onuki: unpublished.
[19] K. Yamaji: J. Phys. Soc. Jpn. 58 (1989) 1520.
[20] M. Nakano: J. Phys. Soc. Jpn. 68 (1999) 1801.
[21] E. Ohmichi, Y. Maeno and T. Ishiguro: J. Phys. Soc. Jpn. 68 (1999) 24.
[22] H. Shibata and H. Fukuyama: J. Phys. Soc. Jpn. 26 (1969) 910.
[23] R. W. Stark and R. Reifenberger: J. Low Temp. Phys. 26 (1977) 763.
[24] Y. Yoshida, R. Settai, Y. Onuki, H. Takei, K. Betsuyaku and H. Harima: J. Phys. Soc. Jpn. 67 (1998) 1677.