Quantum oscillations of magnetization in tight-binding electrons on a honeycomb lattice

Keita Kishigi
Faculty of Education, Kumamoto University, Kurokami 2-40-1, Kumamoto 860-8555, Japan

Yasumasa Hasegawa
Department of Material Science, Graduate School of Material Science, University of Hyogo, Hyogo 678-1297, Japan
(Received 27 January 2014; revised manuscript received 24 June 2014; published 22 August 2014)

We show that quantum oscillations of the magnetization can occur when the Fermi surface consists of points (massless Dirac points) or even when the chemical potential is in an energy gap by studying tight-binding electrons on a honeycomb lattice in a uniform magnetic field. The quantum oscillations of the magnetization as a function of the inverse magnetic field are known as de Haas–van Alphen (dHvA) oscillations and the frequency is proportional to the area of the Fermi surface. The dominant period of the oscillations shown in this paper corresponds to the area of the first Brillouin zone and its phase is zero. The origin of these quantum oscillations is the characteristic magnetic field dependence of the energy known as the Hofstadter butterfly and the Harper broadening of Landau levels. These oscillations are not caused by the crossing of the chemical potential and Landau levels, which is the case in dHvA oscillations. These oscillations can be observed experimentally in systems with a large supercell such as a graphene antidot lattice or ultracold atoms in an optical lattice at an external magnetic field of a few Tesla when the area of the supercell is 10^4 times larger than that of graphene.

DOI: 10.1103/PhysRevB.90.085427 PACS number(s): 71.18.+y, 71.20.−b, 71.70.Di, 81.05.ue

I. INTRODUCTION

Shubnikov–de Haas (SdH) and de Haas–van Alphen (dHvA) oscillations are powerful tools for observing the Fermi surface in conductors [1]. The magnetoresistance for the SdH and the magnetization for the dHvA oscillate periodically as a function of the inverse of a magnetic field (H), respectively. The extremal area of the Fermi surface in a plane perpendicular to the magnetic field is obtained from the period of the oscillations. In a semiclassical approximation the energy of electrons in two-dimensional systems is quantized into Landau levels ε_n due to a uniform magnetic field as

$$F(\varepsilon_n) = (n+\gamma)\frac{2\pi eH}{\hbar c}, \tag{1}$$

where $F(\varepsilon_n)$ is an area of the Fermi surface for $H = 0$ in the wave-number space with the chemical potential $\mu = \varepsilon_n$, n is the Landau index, e is the electron charge, c is the speed of light, $\hbar$ is the Planck constant divided by 2π, and γ is the phase factor. For normal electrons, $\gamma = 1/2$. For massless Dirac fermions, $\gamma = 0$, which are realized in graphene [2] and α-(BEDT-TTF)$_2$I$_3$ [3]. For normal electrons with an effective mass m, the Landau levels are quantized as

$$\varepsilon_n^{\text{normal}} = \frac{e\hbar}{mc}\left(n+\frac{1}{2}\right)H, \tag{2}$$

and the oscillatory part of the magnetization with the constant μ is given by the generalized Lifshitz and Kosevich (LK) formula [1,4–9],

$$M^{\text{LK}} = -\frac{e}{2\pi^2 c\hbar}\frac{F}{\frac{\partial F}{\partial \mu}}\sum_{l=1}^{\infty}\frac{1}{l}R_T^{(l)}\sin\left[2\pi l\left(\frac{f}{H}+\gamma\right)\right], \tag{3}$$

where $R_T^{(l)} = \frac{\lambda l}{\sinh \lambda l}$ is the temperature reduction factor for the lth harmonic, $\lambda = \frac{2\pi^2 k_B T}{\hbar\omega_c}$, k_B is the Boltzmann constant, T is the temperature, and $\omega_c = eH/(cm)$ is the cyclotron frequency. Due to the crossing of μ and Landau levels the magnetization oscillates periodically as a function of $1/H$ with a frequency

$$f = \frac{c\hbar F}{2\pi e}. \tag{4}$$

We have not taken account the effects of Zeeman splitting and impurities. The magnetization changes in a so-called inverse sawtooth pattern as a function of $1/H$, since the coefficient of the lth harmonics is proportional to $1/l$ at $T = 0$. When the number of electrons instead of the chemical potential is fixed, the chemical potential also oscillates and the sawtooth pattern is inverted. Recently the LK formula was shown to be applicable for Dirac electrons in the case of small μ if we use the appropriate n and H dependences of the Landau levels and cyclotron frequency [7–9].

In the LK formula, however, the broadening of Landau levels due to the periodic potentials or the tight-binding nature of the electrons is not taken into account. The broadening of Landau levels is known as Harper broadening [10,11], which makes the Hofstadter butterfly diagram [10]. When the magnetic flux through the unit cell is p/q times the flux quantum $\phi_0 = 2\pi\hbar c/e$, where p and q are mutually prime integers, Landau levels split into p bands in the weak periodic potential case and the Bloch band splits into q bands in the tight-binding model with one orbit in a unit cell and into $2q$ bands in the tight-binding model on a honeycomb lattice. Hall conductance is quantized when μ is in the rth gap and it is given by the Chern number t_r obtained from the Diophantine equation $r = qs_r + pt_r$ [12–14]. The total energy of the electrons is minimized when a magnetic field with one flux quantum per each electron is applied [15,16]. Recently, Hofstadter butterfly diagrams have been observed experimentally in ultracold atoms in optical lattices [17,18] and moiré superlattices [19]. In a graphene antidot lattice [20], the energy band obtained is similar to the Hofstadter butterfly diagram.

The magnetization in a tight-binding model has been studied by many authors [21–29]. In previous studies the oscillations of the magnetization are thought to be caused by the crossing of μ and Landau levels (i.e., the dHvA oscillations) [21–29]. Gat and Avron [25,26] have shown analytically, as a function of μ, the magnetization near commensurate magnetic fluxes in the semiclassical approximation in a square lattice. They have shown that besides the dHvA oscillations, which is zero when μ is at the center of the gap, the mean magnetization has contributions from the Berry phase and the Wilkinson-Rammal (WR) phase [26]. As we will show below, the oscillations of magnetization exist even when μ is fixed in the middle of the gap (namely, μ does not cross the Landau levels), where the quantized Hall conductance is zero. Thus it is not clear whether or not these oscillations are caused by Berry or WR phases. Taut *et al.* [27] reported rapid oscillations of the magnetization numerically in the tight-binding model on a square lattice, in addition to the dHvA oscillations, where there is always a Fermi surface at $H = 0$. In this paper we study the total energy and the magnetization as a function of H for $\mu = 0$ in tight-binding electrons on a honeycomb lattice.

II. MODEL

The Hamiltonian of tight-binding electrons with nearest-neighbor hoppings in a magnetic field is given by

$$\mathcal{H} = -\sum_{(i,j)} t_{i,j} e^{i\theta_{ij}} c_i^\dagger c_j + \varepsilon_A \sum_{i\in A} c_i^\dagger c_i + \varepsilon_B \sum_{i\in B} c_i^\dagger c_i, \quad (5)$$

where

$$\theta_{ij} = \frac{2\pi}{\phi_0} \int_{\mathbf{r}_i}^{\mathbf{r}_j} \mathbf{A}\cdot d\boldsymbol{\ell} \quad (6)$$

and $\mathbf{A}$ is the vector potential. The hopping integrals $t_{i,j}$ between site $\mathbf{r}_i$ and $\mathbf{r}_j$ are $t = 1$, when $\mathbf{r}_i$ and $\mathbf{r}_j$ are the nearest sites and otherwise $t_{i,j} = 0$. We have introduced the site energies ε_A and ε_B for the A and B sublattices. When $H = 0$ and $\varepsilon_A = \varepsilon_B$, the Fermi surface in the half-filled case consists of two Dirac points. When the inversion symmetry is broken by a different potential at each sublattice, a finite gap is opened. We study the cases when the magnetic field $\mathbf{H} = \nabla \times \mathbf{A}$ is uniform and the flux through the unit cell $\phi = \sqrt{3}Ha^2/2$, where a is a lattice constant, is taken as a rational number,

$$\frac{\phi}{\phi_0} = \frac{p}{q} \equiv h. \quad (7)$$

Hereafter, we use h instead of H. The energy is obtained as the eigenvalues $\varepsilon_{i,\mathbf{k}}$ of the $2q \times 2q$ matrix for each wave number $\mathbf{k} = (k_x, k_y)$. The thermodynamic potential per site (Ω), the total energy per site at $T = 0$ (E), and the magnetization at $T = 0$ (M) are calculated by

$$\Omega = -\frac{k_B T}{2qN} \sum_{i=1}^{2q} \sum_{\mathbf{k}} \log\left\{\exp\left(\frac{\mu - \varepsilon_{i,\mathbf{k}}}{k_B T}\right) + 1\right\}, \quad (8)$$

$$E = \frac{1}{2qN} \sum_{\varepsilon_{i,\mathbf{k}} \leqslant \mu} (\varepsilon_{i,\mathbf{k}} - \mu), \quad (9)$$

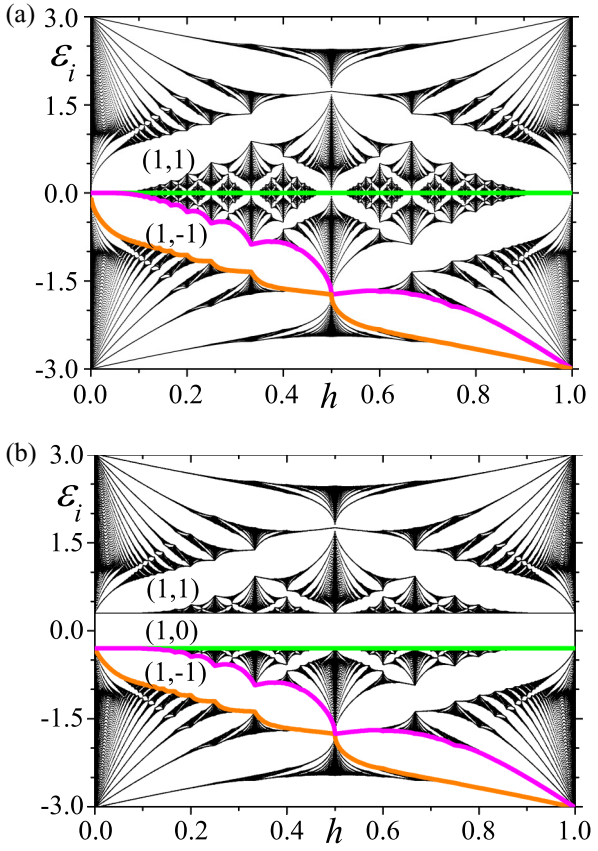

FIG. 1. (Color online) Hofstadter butterfly diagrams of tight-binding electrons on a honeycomb lattice (a) and that with the difference between sublattices A and B ($\varepsilon_A = -\varepsilon_B = 0.3$) (b). In the half-filled case the chemical potential is $\mu = 0$ independent of h. In (b), μ is in the gap. (1,1), (1,0), and (1,−1) are the indices of the gap (s_r, t_r).

and

$$M = -\frac{\partial E}{\partial h}, \quad (10)$$

respectively, where N is the number of points of $\mathbf{k}$. Since the eigenvalues depend on $\mathbf{k}$, there are $2q$ energy bands. This property is different from the semiclassical quantization, where Landau levels are treated as delta functions. For sufficiently large q (e.g., $q = 907$), however, the width of each band is narrow and we can represent the energy for each band at fixed $\mathbf{k}$ (e.g., $\mathbf{k} = \mathbf{0}$). Therefore, $N = 1$ and we obtain the energy of each band as ε_i ($i = 1, 2, \ldots, 2q$) as a function of h. Then we obtain the Hofstadter butterfly diagram as shown in Fig. 1.

III. RESULTS AND DISCUSSION

The total energy is a continuous function of $1/h$ but it has many dips, as shown in Fig. 2, in which we plot total energies for the half-filled ($\mu = 0$) case as a function of $1/h$ for several values of $\varepsilon_A = -\varepsilon_B$. We calculate the magnetization by numerical differentiation by changing p with fixed $q = 907$. Essentially the same results are obtained even if we change the value of q (for example, $q = 499, 467,$

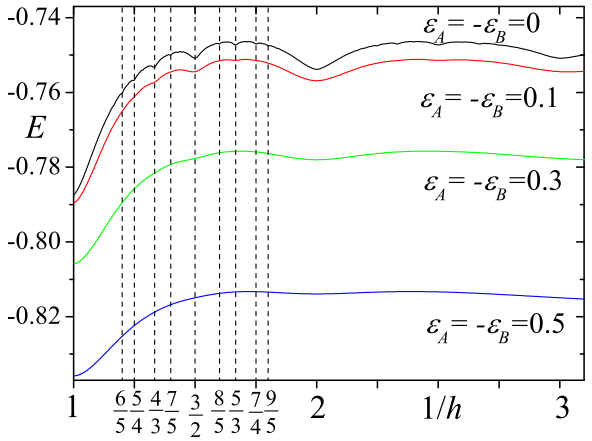

FIG. 2. (Color online) Total energies as a function of $1/h$ for some on-site potentials.

etc). Therefore, the numerical errors due to the differentiation instead of the derivative are expected to be negligibly small. The magnetization at $T = 0$ as a function of $1/h$ is shown in Fig. 3 for $\varepsilon_A = -\varepsilon_B = 0$, 0.1, 0.3, and 0.5.

In the case of $\varepsilon_A = -\varepsilon_B = 0$, the Fermi surface at $h = 0$ is two Dirac points. When $\varepsilon_A = -\varepsilon_B \neq 0$, there is an energy gap between two bands at $h = 0$ and the chemical potential is in the gap, i.e., there is no Fermi surface. In the generalized LK formula [Eq. (3)], the magnetization

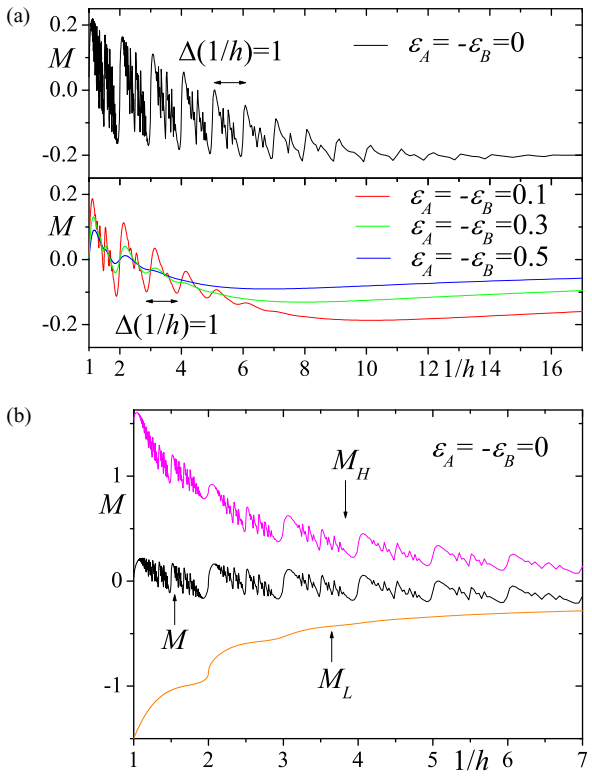

FIG. 3. (Color online) Magnetization of the half-filled case of tight-binding electrons on a honeycomb lattice as a function of h. In (b) contributions from the $n = 0$ band (M_H) and others (M_L) are plotted separately.

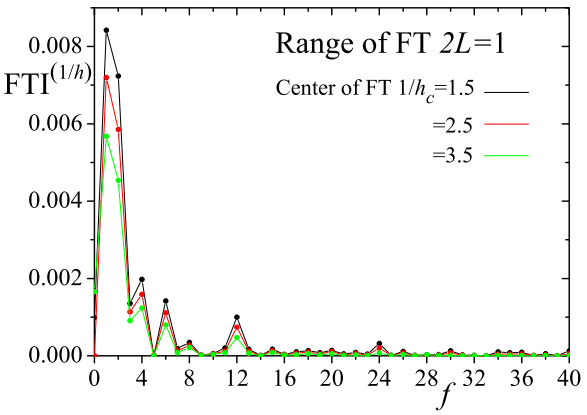

FIG. 4. (Color online) FTIs for $\varepsilon_A = -\varepsilon_B = 0$ of Fig. 3(a).

oscillates with the frequency f, which is proportional to the area of the Fermi surface [Eq. (4)]. Therefore, the oscillations of the magnetization are not expected in half-filled tight-binding electrons on a honeycomb lattice. However, as shown in Fig. 3(a), the magnetization oscillates, although the oscillations of the magnetization on $1/h$ are not perfectly periodic. The amplitudes of the oscillations decrease as $1/h$ increases. The shape is not a perfect sawtooth, but it is similar to the sawtooth pattern for fixed electron numbers rather than the inverse sawtooth pattern for the fixed chemical potential, although it is chaotic for small $1/h$. The most dominant period of the oscillations of M as a function of $1/h$ is 1, which corresponds to the area of the first Brillouin zone in the LK formula. The phase of the oscillations corresponds to zero ($\gamma = 0$) in the LK formula. Since the magnetization is not a perfect periodic function of $1/h$, we calculate the Fourier transform, choosing the center $1/h_c$ and a finite range $2L$, as

$$\mathrm{FTI}^{(1/h)}\left(f, \frac{1}{h_c}, L\right) = \left| \frac{1}{2L} \int_{\frac{1}{h_c}-L}^{\frac{1}{h_c}+L} M(h) e^{2\pi i \frac{f}{h}} d\left(\frac{1}{h}\right) \right|^2 . \tag{11}$$

Since we perform the Fourier transform in the finite range $2L$, we take $f = j/(2L)$ with integer j. We plot the Fourier transform intensities (FTIs) for $\varepsilon_A = -\varepsilon_B = 0$, $2L = 1$, and $1/h_c = 1.5$, 2.5, and 3.5 in Fig. 4. If the oscillations would have a perfect sawtooth pattern, FTIs would decrease as $1/f^2$ and not depend on the choice of $1/h_c$. As shown in Fig. 4, the components of $f = 1$,2, 4, 6, 12, 24, etc., are large and those of $f = 5$,7, 9, etc., are small. The amplitudes become small when we take larger $1/h_c$, but the f dependences are similar.

In order to see the magnetization as a function of $1/h$ in detail, we plot M for $1 \leqslant 1/h \leqslant 2$ in Fig. 5. There are many jumplike structures when $1/h$ is a rational number ($1/h = q/p$) written by small integers (p and q). However, it is clearly seen that they are not discontinuous jumps but continuous sharp cliffs at $1/h = 1$, 4/3, 3/2, 5/3, 2, etc. There are large sawtoothlike oscillations with periods of 1, $\frac{1}{2}$, $\frac{1}{6}$, and $\frac{1}{12}$, which contribute to the peaks of $\mathrm{FTI}^{(1/h)}(f, 1/h_c, L)$ for $f = 1$, 2, 4, 6, 12, and 24.

In order to clarify the origin of the magnetic field dependence of the magnetization, we calculate the contributions

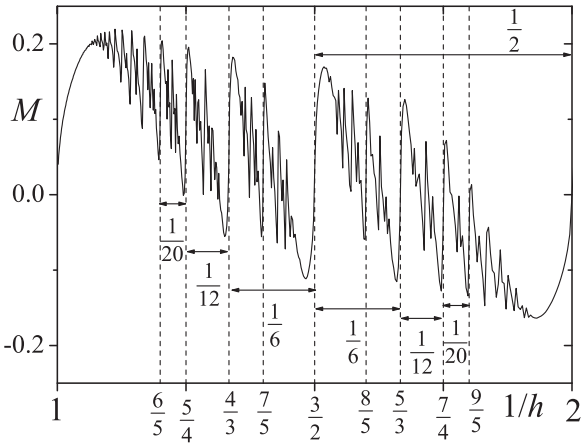

FIG. 5. Closeup of Fig. 3(a) for $\varepsilon_A = -\varepsilon_B = 0$ (magnetization of the half-filled case of tight-binding electrons on a honeycomb lattice as a function of $1/h$).

of the different parts of the Hofstadter butterfly diagram separately. We define E_H, E_L, M_H, and M_L as

$$E_H = D \sum_{\varepsilon_n \in R_H} \varepsilon_n, \tag{12}$$

$$E_L = D \sum_{\varepsilon_n \in R_L} \varepsilon_n, \tag{13}$$

$$M_H = -\frac{\partial E_H}{\partial h}, \tag{14}$$

and

$$M_L = -\frac{\partial E_L}{\partial h}, \tag{15}$$

where R_H is the set of eigenstates with the energy between $\varepsilon_i = 0$ and a large gap starting from $\varepsilon_i = 0$ at $h = 0$ [states between the green and pink curves in Fig. 1(a)], and R_L is a set of eigenstates with the energy below the large gap [states below the orange curve in Fig. 1(a)]. In Fig. 3(b) we plot M_H, M_L, and $M = M_H + M_L$ as functions of $1/h$ for $\varepsilon_A = -\varepsilon_B = 0$. The oscillatory dependence of the magnetization comes from M_H. The states between the green and pink curves in Fig. 1(a) are the $n = 0$ Landau levels in the continuum limit. Namely, the oscillations of M_H in Fig. 3(b) are thought to be caused by the broadening of Landau levels due to tight-binding electrons.

Next, we study the temperature dependence of the FTIs, which are shown in Fig. 6. Although the LK formula cannot be applied in the present case ($R_T^{(l)}$ is always 1, if we set the effective mass to be zero), we try to fit the temperature reduction as the reduction factor for normal electrons with the effective mass. We see that the effect of the temperature can be fitted by the effective mass $m = 2.5$, where the unit of the mass is $\hbar^2/(ta^2)$, at a low temperature region ($T \lesssim 0.04$), where the unit of T is t, but it deviates as the temperature becomes large.

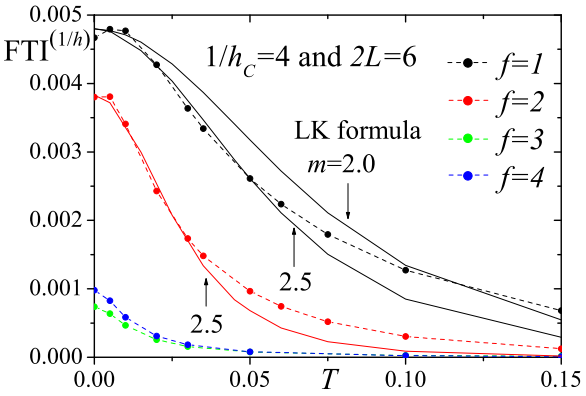

FIG. 6. (Color online) T dependences of the FTIs with $f = 1, 2, 3, 4$. The Fourier transformation is done in the region $1 \leqslant 1/h \leqslant 7$. Solid lines are T dependences of the FTIs of the LK formula with $m = 2.0$ and $m = 2.5$. The numerically calculated cyclotron mass at $\mu = -0.8$ in the tight-binding model is about 2.5.

IV. CONCLUSION

We have shown that the oscillations of the magnetization as a function of the magnetic field exist even when the area of the Fermi surface vanishes, if the tight-binding electrons on a honeycomb lattice are studied. Since the chemical potential stays in the energy gap when $\epsilon_A \neq \epsilon_B$, these quantum oscillations are not caused by the crossing of the chemical potential and the energy bands (Landau levels), which is the origin of the dHvA oscillations in the case of a semiclassical approximation. They come from the complex structure of the $n = 0$ Landau level in the Hofstadter butterfly diagram. The origin of these oscillations can also be considered as the simultaneous crossings of Landau levels [$\pm n$ starting from the bottom and top of the energy ($\epsilon = \mp 3$) at $h = 0$], which do not change the energy gap at $\epsilon = 0$. The effect of temperature reduces the amplitudes of the oscillations, but it is not described by the temperature reduction factor in the LK formula. We can fit the temperature dependence by the temperature reduction factor $R_T^{(l)}$ only in a small region of temperature.

It is difficult to observe these quantum oscillations of magnetization experimentally, because $h \simeq 1/16$ means $H \simeq 5000$ T from $a \simeq 0.246$ nm of graphene and $H \simeq 250$ T from the area of unit cell [30] of α-(BEDT-TTF)$_2$I$_3$ [3], respectively. However, if the area of the supercell in graphene antidot lattices [20] or ultracold atoms on the optical lattice [31] is taken to be about 10^4 times larger than that of graphene, these quantum oscillations of the magnetization may be observed at a few Tesla. In this paper, we have neglected the Zeeman energy. When the energy bands for up and down spins are overlapped by the Zeeman splitting, these quantum oscillations will be suppressed. However, it will be possible to make a system have an energy gap ($|\epsilon_A - \epsilon_B|$) at $H = 0$ larger than $\mu_B H$, where μ_B is the Bohr magneton and H is a few Tesla. In that system the chemical potential stays in the energy gap even when the Zeeman energy is taken into account, and it will be possible to observe these oscillations.

[1] D. Schoenberg, *Magnetic Oscillations in Metals* (Cambridge University Press, Cambridge, UK, 1984).
[2] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, M. I. Katsnelson, I. V. Grigorieva, S. V. Dubonos, and A. A. Firsov, Nature (London) **438**, 197 (2005).
[3] N. Tajima, T. Yamauchi, T. Yamaguchi, M. Suda, Y. Kawasugi, H. M. Yamamoto, R. Kato, Y. Nishio, and K. Kajita, Phys. Rev. B **88**, 075315 (2013).
[4] I. M. Lifshitz and A. M. Kosevich, Zh. Eksp. Teor. Fiz. **29**, 730 (1955) [Sov. Phys. JETP **2**, 636 (1956)].
[5] T. Champel, Phys. Rev. B **64**, 054407 (2001).
[6] K. Kishigi and Y. Hasegawa, Phys. Rev. B **65**, 205405 (2002).
[7] I. A. Luk'yanchuk and Y. Kopelevich, Phys. Rev. Lett. **93**, 166402 (2004).
[8] I. A. Luk'yanchuk, Low Temp. Phys. **37**, 45 (2011).
[9] S. G. Sharapov, V. P. Gusynin, and H. Beck, Phys. Rev. B **69**, 075104 (2004).
[10] D. R. Hofstadter, Phys. Rev. B **14**, 2239 (1976).
[11] P. G. Harper, Proc. Phys. Soc., London, Sect. A **68**, 874 (1955).
[12] D. J. Thouless, M. Kohmoto, M. P. Nightingale, and M. den Nijs, Phys. Rev. Lett. **49**, 405 (1982).
[13] Y. Hasegawa and M. Kohmoto, Phys. Rev. B **74**, 155415 (2006).
[14] Y. Hasegawa and M. Kohmoto, Phys. Rev. B **88**, 125426 (2013).
[15] Y. Hasegawa, P. Lederer, T. M. Rice, and P. B. Wiegmann, Phys. Rev. Lett. **63**, 907 (1989).
[16] Y. Hasegawa, Y. Hatsugai, M. Kohmoto, and G. Montambaux, Phys. Rev. B **41**, 9174 (1990).
[17] M. Aidelsburger, M. Atala, M. Lohse, J. T. Barreiro, B. Paredes, and I. Bloch, Phys. Rev. Lett. **111**, 185301 (2013).
[18] H. Miyake, G. A. Siviloglou, C. J. Kennedy, W. C. Burton, and W. Ketterle, Phys. Rev. Lett. **111**, 185302 (2013).
[19] C. R. Dean, L. Wang, P. Maher, C. Forsythe, F. Ghahari, Y. Gao, J. Katoch, M. Ishigami, P. Moon, M. Koshino, T. Taniguchi, K. Watanabe, K. L. Shepard, J. Hone, and P. Kim, Nature (London) **497**, 598 (2013).
[20] J. G. Pedersen and T. G. Pedersen, Phys. Rev. B **87**, 235404 (2013).
[21] K. Machida, K. Kishigi, and Y. Hori, Phys. Rev. B **51**, 8946 (1995).
[22] K. Kishigi, M. Nakano, K. Machida, and Y. Hori, J. Phys. Soc. Jpn. **64**, 3043 (1995).
[23] P. S. Sandhu, J. H. Kim, and J. S. Brooks, Phys Rev. B **56**, 11566 (1997).
[24] S. Y. Han, J. S. Brooks, and J. H. Kim, Phys. Rev. Lett. **85**, 1500 (2000).
[25] O. Gat and J. E. Avron, New J. Phys. **5**, 44 (2003).
[26] O. Gat and J. E. Avron, Phys. Rev. Lett. **91**, 186801 (2003).
[27] M. Taut, H. Eschrig, and M. Richter, Phys. Rev. B **72**, 165304 (2005).
[28] V. M. Gvozdikov and M. Taut, Phys. Rev. B **75**, 155436 (2007).
[29] W. H. Xu, L. P. Yang, M. P. Qin, and T. Xiang, Phys. Rev. B **78**, 241102 (2008).
[30] K. Bender, I. Hennig, D. Schweitzer, K. Dietz, H. Endres, and H. J. Keller, Mol. Cryst. Liq. Cryst. **108**, 359 (1984).
[31] L. Tarruell, D. Greif, T. Uehlinger, G. Jotzu, T. Esslinger, and J. L. McChesney, Nature (London) **483**, 302 (2012).