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Ground state magnetization of conduction electrons in graphene with Zeeman effect

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A B S T R A C T

In this work we address the ground state magnetization in graphene, considering the Zeeman effect and taking into account the conduction electrons in the long wavelength approximation. We obtain analytical expressions for the magnetization at T=0 K, where the oscillations given by the de Haas van Alphen (dHvA) effect are present. We find that the Zeeman effect modifies the magnetization by introducing new peaks associated with the spin splitting of the Landau levels. These peaks are very small for typical carrier densities in graphene, but become more important for higher densities. The obtained results provide insight of the way in which the Zeeman effect modifies the magnetization, which can be useful to control and manipulate the spin degrees of freedom.

1. Introduction

Since its experimental isolation in 2004, graphene has become one of the most studied and promising material in condensed matter physics [1–4]. Its interesting properties are related with its 2D hexagonal structure, made of two interpenetrating sublattices A and B which behave as a pseudospin degrees of freedom [5]. Without impurities or defects, the conduction and valence bands touch at the Fermi energy, with the valence band full and the conduction band empty in the ground state [4]. Furthermore, in pristine graphene the density of states at the Fermi energy is zero, and thus the graphene is a semiconductor with zero band gap, or a semi-metal [6]. In the long wavelength approximation the dispersion relation is relativistic and the electrons behave as massless fermions, moving with a Fermi velocity of about c/300 [7].

When a magnetic field is applied to graphene, discrete Landau levels are obtained [9]. For a classical electron gas these levels are equidistant, due to a parabolic dispersion relation. For a relativistic-like electron gas, like in graphene, the Landau levels are not equidistant, which is one of the reasons quantum Hall effect can be observed in graphene at room temperatures [10–15]. Moreover, Landau levels create an oscillating behavior in the thermodynamics potentials. It is found that the magnetization oscillates as a function of the inverse magnetic field, the so called de Haas van Alphen effect [18,19]. The different frequencies involved in the oscillations are related to the closed orbits that electrons perform on the Fermi surface [20]. It has been predicted, in graphene, that the magnetization oscillates periodically in a sawtooth pattern, in agreement with the old Peierls prediction [21]. In contrast to 2D conventional semiconductors, where the oscillating center of the magnetization is zero, in graphene the oscillating center has a positive value because the diamagnetic contribution is half reduced with that in the conventional semiconductor [22].

When we consider the Zeeman effect, the Landau levels for each spin split introducing a gap. This splitting becomes relevant when the thermodynamical properties are considered [23,24]. Indeed, the splitting affects the filling of the energy states when the internal energy is calculated, and consequently other related functions such as the magnetization. In general, the parameters that affect the occupancy of the energy levels are the electron density n_e and the magnetic field. Thus one can conceive a graphene-like system with its valence band fully occupied and only the conduction band available, in such a way that n_e can be modified. The added electrons could be originated by a gate voltage V_G applied to the graphene sheet so that n_e can be varied as a function of V_G. This system may be found useful in the characterization of spin-filter [25] and spin-polarized currents in 2D systems [26], which in turn can be used to calculate transport parameters like charge and spin conductivity [27]. Motivated by this facts we studied the magnetization at T=0 K in a general graphene-like system with only the conduction band available and n_e variable, taking into account the Zeeman effect and the way in which the magnetic oscillations are altered by this effect.

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2. Graphene in magnetic field

2.1. Theoretical framework

We considered the conduction electrons in our graphene system in the long wavelength approximation, which implies low energies such that $E \ll t$~3 eV (where t is the NN hopping amplitude [4]). This gives a relativistic-like dispersion relation $E=\hbar v_F|\mathbf{k}|$, where v_F~10^6 m/s is the Fermi velocity. We suppose that the conduction electrons have an electron density n_e, which may be due to an applied gate voltage. The long wavelength approximation is valid if n_e is such that the Fermi energy E_F obeys $E_F \ll t$~3 eV. If we have N conduction electrons in an area A, then $n_e = N/A$, and the density of states in the long wavelength approximation is $\rho(E)=d2\pi AE/h^2v_F^2$, where d=4 takes into account the spin and valley degeneracy. Thus

$$N = \int_0^{E_F} \frac{8\pi AE}{h^2 v_F^2} dE. \tag{1}$$

Therefore the condition $E_F \ll t$ implies

$$n_e \ll \frac{4\pi t^2}{h^2 v_F^2}. \tag{2}$$

For t~3 eV, Eq. (2) is satisfied for typical carrier densities in graphene [28–31] (about $n_e<10^{12}c\mathrm{m}^{-2}$). Then we shall take the regime $n_e \leq 0.\ 1n\mathrm{m}^{-2}$.

The graphene Hamiltonian in the long wavelength approximation is[1]

$$H = v_F(\boldsymbol{\sigma}\cdot\mathbf{p}), \tag{3}$$

where $\boldsymbol{\sigma}=(\sigma_x, \sigma_y)$ are the Pauli matrices, which act in the sublattices A and B of graphene. Applying a magnetic field, the momentum changes following the Peierls substitution [32] $\mathbf{p} \to \mathbf{p} - e\mathbf{A}$, where $\mathbf{A}$ is the vector potential. For a magnetic field $\mathbf{B} = (0, 0, B)$, in the Landau gauge we have $\mathbf{A}=(-By,0, 0)$. Considering the Zeeman effect [33], the term $\boldsymbol{\mu}\cdot\mathbf{B} = \mu_B g B s_z/2$ is added to H, where $s_z=2S_z/\hbar$ is the Pauli matrix acting in the spin state and g≃2. Therefore Eq. (3) now reads

$$H = v_F[\sigma_x(p_x + eBy) + \sigma_y p_y] - \boldsymbol{\mu}\cdot\mathbf{B}. \tag{4}$$

Because H only depends on the y coordinate, then we can express the wave function as $\psi = e^{-ikx}(\psi^A \quad \psi^B)$, with $\psi^{A/B}$ depending only on y. Replacing $p_i=-i\hbar\partial_i$ in Eq. (4), the equation $H\psi = E\psi$ becomes

$$[v_F(\sigma_x(-\hbar k + eBy) - i\hbar\sigma_y\partial_y) - \boldsymbol{\mu}\cdot\mathbf{B}]\psi = E\psi. \tag{5}$$

Introducing the ladder matrices $\sigma_\pm = \sigma_x \pm i\sigma_y$ and making the change of variable $y'=(-\hbar k + eBy)/\sqrt{\hbar eB}$ [34] we can write Eq. (5) as

$$\left[v_F\sqrt{\hbar eB}\,\frac{\sigma_+}{2}(y'-\partial_{y'}) + v_F\sqrt{\hbar eB}\,\frac{\sigma_-}{2}(y'+\partial_{y'}) - \boldsymbol{\mu}\cdot\mathbf{B}\right]\psi = E\psi. \tag{6}$$

This Hamiltonian is identical to the quantum harmonic oscillator. Indeed, defining the ladders operators $a^\dagger = (y'-\partial_{y'})/\sqrt{2}$ and $a = (y'+\partial_{y'})/\sqrt{2}$ we have

$$\left[\frac{\hbar\omega_L}{2}(\sigma_+ a^\dagger + \sigma_- a) - \hbar\omega_Z s_z\right]\psi = E\psi, \tag{7}$$

where $\omega_L = v_F\sqrt{\frac{2eB}{\hbar}}$ and $\omega_Z = \mu_B B/\hbar$. The energies from Eq. (7) can be calculated by writing the wave function as

$$|\psi\rangle = c_1|n, A,+\rangle + c_2|n-1,B,+\rangle + c_3|n, A,-\rangle + c_4|n-1,B,-\rangle, \tag{8}$$

where $|+\rangle$ and $|-\rangle$ represent spin up and down, so that $s_z|\pm\rangle=\pm|\pm\rangle$. Then, given that $\sigma_+|A\rangle=0$, $\sigma_+|B\rangle=2|A\rangle$, $\sigma_-|A\rangle=2|B\rangle$, $\sigma_-|B\rangle=0$ and $a^\dagger|n\rangle = \sqrt{n+1}\,|n+1\rangle$, $a|n\rangle = \sqrt{n}\,|n-1\rangle$, solving Eq. (7) the energies are

$$E_{n,s,l} = l\hbar\omega_L\sqrt{n} - s\hbar\omega_Z, \tag{9}$$

where n=0, 1, 2… is the Landau level index, s= ± 1 for spin up and down and l= ± 1 for the valence and conduction band. For the K' valley the energies are identical to Eq. (9), so that each state is doubly degenerate. The Zeeman interaction splits the spin up and down energies, introducing a gap given by $\Delta E=2\hbar\omega_Z$. As in the classical case, the degeneracy of each spin level is given by $D=2AB/(h/e)=AB/\phi$, where A is the area of graphene sheet and $\phi = h/(2e)$ is half the magnetic unit flux [7].[2]

To study the ground state magnetization we considered that only the conduction band is available. The valence band, although full in our model, would still make a continuous non oscillatory contribution to the magnetization. Since we are interested only in the magnetic oscillations, we shall omit the valence band and work only with the conduction electrons. Thus the energies are $\varepsilon_n^s=\hbar\omega_L\sqrt{n} - s\hbar\omega_Z$, where s= ± 1 for spin up and down. The internal energy for N electrons can be computed as the sum of the filled Landau levels. The number of totally filled levels is $q = [q_c]$, where $q_c = N/D$ is the filling factor, and the brackets means the biggest integer less or equal to q_c (the Floor function). We can also write $N/D = B_C/B$ where $B_C = n_e\phi$ ($n_e = N/A$) is the critical magnetic field at which the degeneracy D equals the number of electrons N.

In order to calculate the internal energy we first have to sort the energy levels. It may happen that the splitting is such that for a given Landau level n, $\varepsilon_{n+1}^+ < \varepsilon_n^-$, which would mean that the states with energy ε_{n+1}^+ are filled before those with energy ε_n^-. This would happen if $\hbar\omega_L(\sqrt{n+1} - \sqrt{n})<2\hbar\omega_Z$. For q levels filled, considering that each state may be occupied with spin up or down, the condition at which the mixing starts can be approximated by

$$\hbar\omega_L\left(\sqrt{\frac{q}{2}} - \sqrt{\frac{q}{2}-1}\right)<2\hbar\omega_Z. \tag{10}$$

In general this condition depends on the electron density n_e because $q = [n_e\phi/B]$. Nevertheless, it can be easily proved that Eq. (10) occurs only for electron densities that do not satisfy Eq. (2). Therefore there is no spin mixing at the long wavelength approximation with magnetic field. In order to study the spin mixing, one would have to take in consideration the whole dispersion relation of Bloch electrons in graphene [7], in which case the problem becomes increasingly difficult [8].

2.2. Ground state magnetization

We call ξ_m the decreasing sorted energy levels, m being the label index. We can write $\xi_m = \varepsilon_m^0 - (-1)^m\beta\hbar\omega_Z$, where $\varepsilon_m^0=\hbar\omega_L[\frac{m}{2} - \frac{1}{4}(1-(-1)^m)]^{\frac{1}{2}}$ are the Landau levels, written in such a way to ensure that for each Landau level we take both spins. We introduced a parameter β to differentiate the situations without Zeeman effect (β=0) and with Zeeman effect (β=1).

If we call $\theta = q_c - q = N/D - [N/D]$ the occupancy factor of the last partially filled Landau level, the ground-state internal energy is

$$U = \sum_{m=0}^{q-1} D\xi_m + D\theta\xi_q. \tag{11}$$

Replacing the expression for ξ_m we have $U = D\sum_{m=0}^{q-1}\varepsilon_m^0 - \beta D\hbar\omega_Z\sum_{m=0}^{q-1}(-1)^m + D\theta\varepsilon_q^0 - \beta D\hbar\omega_Z(-1)^q$. The factor $\sum_{m=0}^{q-1}(-1)^m$ is 0 if q is even, or 1 if q is odd. Thus we can write $\sum_{m=0}^{q-1}(-1)^m = [1-(-1)^q]/2$. Moreover, the term $U_0=D\sum_{m=0}^{q-1}\varepsilon_m^0+D\theta\varepsilon_q^0$ is the energy without Zeeman effect. Therefore

$$U = U_0 - \beta\frac{1}{2}D\hbar\omega_Z[1+(-1)^q(2\theta-1)]. \tag{12}$$

The last term in Eq. (12) is associated with the spin magnetization.

[1] To find the energies we consider the K valley.

[2] The factor of 2 in D takes into account the valley degeneracy.

To see this, we start from the Pauli magnetization given by

$$M_P = \mu_B(N_+ - N_-), \tag{13}$$

where N_+ and N_- are the total number of spin up and down, respectively. If q is even, the number of spin up and down states totally occupied are identical, and the last unfilled state is spin up. Then $N_+ - N_- = D\theta$ if q is even. On the other hand, if q is odd there is one unpaired totally filled spin up state and the last unfilled state is spin down. Then $N_+ - N_- = D - D\theta$ if q is odd. Therefore in general we can write $N_+ - N_- = D[1+(-1)^q(2\theta-1)]/2$, and Eq. (13) becomes

$$M_P = \mu_B \frac{D}{2}[1+(-1)^q(2\theta-1)]. \tag{14}$$

Consequently the internal energy of Eq. (12) becomes

$$U = U_0 - \beta B M_P. \tag{15}$$

Thus the energy, and related functions such as the magnetization, are altered by the Zeeman effect through the spin magnetization. The magnetization at T=0 K is $M = -\partial U/\partial B$. From Eq. (15) we have

$$M = M_0 + \beta\left(M_P + B\frac{\partial M_P}{\partial B}\right), \tag{16}$$

where $M_0 = -\partial U_0/\partial B$ is the magnetization without Zeeman effect. From Eq. (14) we get $\partial M_P/\partial B = M_P/B + \mu_B D(-1)^q \partial\theta/\partial B$, with $\partial\theta/\partial B = -N/(DB)$. Therefore Eq. (16) becomes

$$M = M_0 + \beta(2M_P - N\mu_B(-1)^q). \tag{17}$$

Fig. 1 shows the magnetization (17) for an electron density n_e=0. 1nm^{-2} and area A=100nm^2, for the case without Zeeman effect (β=0) and the case with Zeeman effect (β=1). We can see that the magnetization oscillates in agreement with the de Haas van Alphen effect. Moreover, because q is a periodic function with 1/B, therefore M is also a periodic function with 1/B, as can be observed in Fig. 1(b). We also notice that the oscillating center of the magnetization has a positive value, which means that conduction electrons have a ground state paramagnetism. This differs substantially with what happens in the conventional semiconductor 2DEG. Thus the results obtained affords an intuitive explanation of the difference in magnetization between the monolayer graphene and the conventional semiconductor 2DEG.

As it can be seen in Fig. 1 the Zeeman effect introduces a second peak in the magnetization. To understand this unusual behavior we have to analyze in more detail the Eq. (17). Without Zeeman effect we have $M_0 = -\partial U_0/\partial B$, with $U_0 = D\sum_{m=0}^{q-1}\varepsilon_m^0 + D\theta\varepsilon_q^0$. Then, given that $\partial D/\partial B = D/B$, $\partial\varepsilon_m^0/\partial B = (1/2B)(\varepsilon_m^0)$ and $\partial\theta/\partial B = -N/(DB)$, we can write

$$M_0 = \frac{1}{B}\left(N\varepsilon_q^0 - \frac{3}{2}U_0\right), \tag{18}$$

and therefore from Eq. (17) the magnetization with Zeeman effect becomes

$$M_Z = \frac{1}{B}\left(N\varepsilon_q^0 - \frac{3}{2}U_0\right) + 2M_P - N\mu_B(-1)^q. \tag{19}$$

From Eq. (18) we see that the peaks appear at M_0 whenever ε_q^0 changes discontinuously, U_0 being continuous. This happens only when q changes from odd to even $\left(\text{recall that } \varepsilon_m^0 = \hbar\omega_L[\frac{m}{2} - \frac{1}{4}(1-(-1)^m)]^{\frac{1}{2}}\right)$, which corresponds to a change of Landau level. On the other hand, Eq. (19) gives peaks in M_z whenever q change because of the additional factor $N\mu_B(-1)^q$; the new peaks are produced by the change of spin. These results imply that without Zeeman effect there is a jump in the magnetization only when the last state changes the Landau level, while with Zeeman effect there is a jump when the last state changes either its spin or Landau level. This effect can also be related to fractional filling factors. To see this consider the energy degeneracy D_L of each Landau level with no Zeeman effect, which can be occupied with spin up or down, so $D_L = 2D = 2AB/\phi$. Then, $q = [N/D] = [2N/D_L] = 2q_L$, where $q_L = [N/D_L]$. For the case with Zeeman effect, the change of spin is associated with q odd, while the change of Landau level with q even. In terms of q_L this implies that the peaks in M given by a change of Landau level correspond to q_L integer, whereas the peaks given by change of spin correspond to q_L fractional. In this way we can say that the peaks produced by the Zeeman effect correspond to fractional filling factors in the case without Zeeman effect. Such behavior is similar to the Fractional Quantum Hall effect in graphene [15–17], where changes appear in the Hall conductivity for fractional occupancy number due to the Coulomb interaction between electrons.

In the case of Zeeman effect, Fig. 1 also shows that the amplitude of the peaks corresponding to a change of spin is smaller than the amplitude corresponding to a change of Landau level.[3] In fact, the amplitude depends on the density of electrons n_e, as can be seen in Eq. (19). If the peak corresponds to a change of spin we have $\Delta M^S = 2N\mu_B = 2A\mu_B n_e$, whereas if it is due to a change in the Landau level we obtain $\Delta M^L = \frac{An_e}{B}\left[\hbar\omega_L\left(\sqrt{\frac{q}{2}} - \sqrt{\frac{q}{2}-1}\right) - 2\hbar\omega_Z\right]$ with q an even integer.

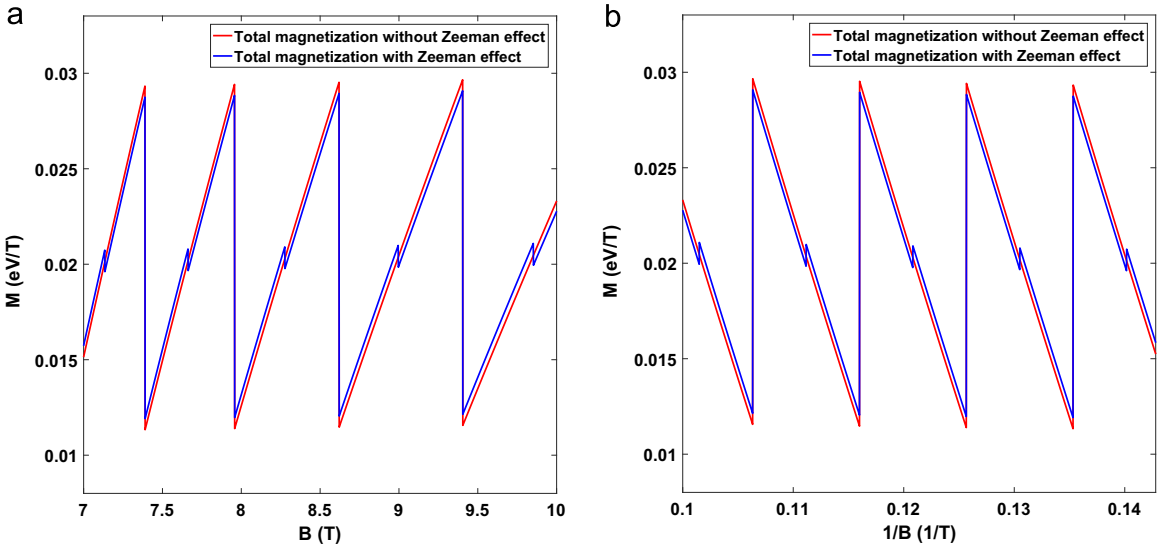

Fig. 1. (a) Ground state magnetization M as a function of B, with and without Zeeman effect. (b) Ground state magnetization M as a function of 1/B, with and without Zeeman effect. In both cases the density of electrons is n_e=0. 1nm^{-2} and the area A=100nm^2.

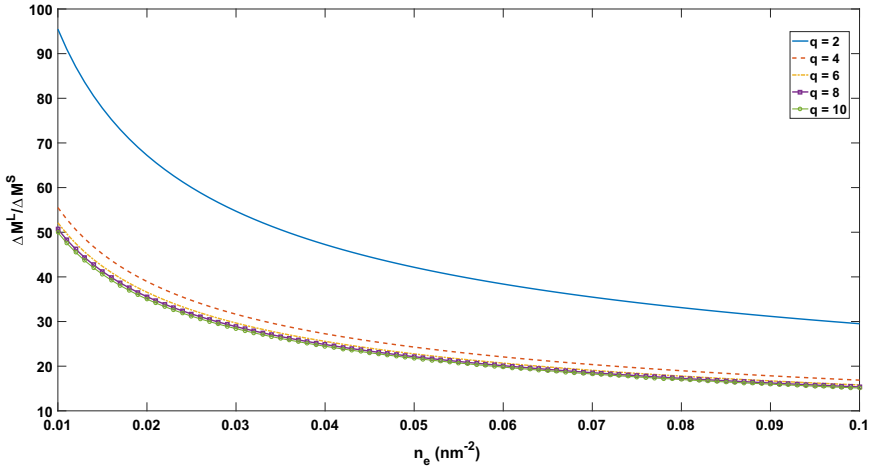

Fig. 2. $\Delta M^L/\Delta M^S$ as a function of n_e, as given by Eq. (20), for different values of $q = n_e\phi/B$ ($\phi = h/2e$).

Notice that ΔM^S depends only on n_e, whereas ΔM^L depends on both n_e and the magnetic field B. Moreover, the spin splitting appears as a reduction factor in the amplitude ΔM^L, as expected [35]. The ratio $\Delta M^L/\Delta M^S$ is

$$\frac{\Delta M^L}{\Delta M^S} = \frac{\hbar\omega_L}{2\hbar\omega_Z}\left(\sqrt{\frac{q}{2}} - \sqrt{\frac{q}{2}-1}\right) - 1 \ (q \ \text{ even integer}) \tag{20}$$

where $q = n_e\phi/B$ ($\phi = h/2e$). In Fig. 2 Eq. (20) is plotted as a function of n_e, for different values of q even. We see that for $n_e \lesssim 0.\ 1n\text{m}^{-2}$ we always have $\Delta M^L > \Delta M^S$, but $\Delta M^L/\Delta M^S$ decreases as n_e increases. For typical carrier densities in graphene, about $n_e < 10^{12}c\text{m}^{-2}$, we have $\Delta M^L \gg \Delta M^S$. This would explain why this phenomenon has not yet been seen experimentally in graphene. Nevertheless, for higher electron densities the effect would be prominent (see Fig. 1). These results are in concordance with [36], where the spin splitting appears as a reduction factor in the magnetic oscillations for 2D normal systems. When the Zeeman splitting becomes a half of the the Landau level spacing, the amplitude of oscillation of the fundamental frequency becomes zero. Indeed, Eq. (20) gives $\Delta M^L = 0$ for $q = 2$ if $\hbar\omega_Z = \hbar\omega_L/2$. Nevertheless, given that $q = n_e\phi/B$, this would happen for electron densities n_e that do not satisfy Eq. (2).

From an experimental point of view, the relations of the peaks can be controlled by the applied electric field that controls the carrier concentration, whereas the spin polarization lifetime can be controlled by the applied gate voltage [37]. This can be useful to improve the methods for mapping the Fermi surface by taking into account the Fourier decomposition of the new peaks.

3. Conclusions

We studied the ground state magnetization of conduction electrons in graphene with Zeeman effect. We considered only the conduction electrons in the long wavelength approximation, which was shown to hold for typical carrier densities in graphene. We have derived analytical expressions for the magnetization at $T = 0$ K, with and without Zeeman effect. It was shown that the magnetization has peaks whenever the last energy level changes discontinuously, and its amplitude depends on the electron density. In the case without Zeeman effect these peaks appear only when the last Landau level occupied changes. With Zeeman effect it was shown that new peaks appear in the magnetization, associated with the spin splitting in the Landau levels. These new peaks occur whenever the last state changes only its spin, while the Landau level remains the same. We also studied the ratio of amplitudes between the peaks produced by a change of Landau level (ΔM^L) and the new peaks produced only by a change of spin (ΔM^S). An analytical expression was derived, which shows that $\Delta M^L \gg \Delta M^S$ for typical carrier densities in graphene, about $n_e < 10^{12}c\text{m}^{-2}$. Nevertheless, for higher electron densities, about $n_e \simeq 0.\ 1n\text{m}^2$, the effect should become evident. These new findings can be verified by studying experimentally graphene at very high carrier densities and perpendicular magnetic field. The predicted effect will hopefully help the interpretation of magnetization in experiments.

[3] When there is a change of Landau level the spin also changes. But this is different to what we simply call a change of spin, where the spin changes but the Landau level is the same.

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