Spin splitting in graphene studied by means of tilted magnetic-field experiments

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We have measured the spin splitting in single-layer and bilayer graphene by means of tilted magnetic field experiments. Applying the Lifshitz-Kosevich formula for the spin-induced decrease of the Shubnikov de Haas amplitudes with increasing tilt angle we directly determine the product between the carrier cyclotron mass \( m^* \) and the effective \( g \)-factor \( g^* \) as a function of the charge carrier concentration. Using the cyclotron mass for a single-layer and a bilayer graphene we find an enhanced \( g \)-factor \( g^* = 2.7 \pm 0.2 \) for both systems.

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The half-integer quantum Hall effect in single-layer graphene (SLG) [1][2] and the unconventional quantum Hall effect in bilayer graphene (BLG) [3] reveal spin- and valley-degenerate relativistic Landau levels. Due to the extremely large Landau-level splitting [4][5], completely resolved levels can be observed up to room temperature [6]. However, even at very high perpendicular magnetic fields the Zeeman splitting within one Landau-level is negligible smaller compared to the Landau-level splitting and, more importantly, the Landau-level width generally exceeds the spin-splitting. Exceptionally, the zeroth Landau level in SLG becomes extremely narrow at magnetic fields \( B > 20 \) T [4], which allows an experimental observation of a spin-related gap opening at magnetic fields \( B > 20 \) T [7]. Another observation of a spin degeneracy lifting with an effective \( g \)-factor \( g^* = 2 \) was reported for \( \nu = \pm 4 \), in SLG for magnetic fields \( B > 30 \) T, combined with lifting the valley-degeneracy at \( \nu = \pm 1 \) [8].

In this paper we determine the spin splitting of broadened Landau levels for SLG and BLG by measuring Shubnikov-de Haas oscillations in tilted magnetic fields. This technique allows adjusting the ratio between the spin splitting and the Landau level splitting, by controlling the ratio between a total magnetic field and a component perpendicular to a two-dimensional graphene flake. Using the well-established Lifshitz-Kosevich formula [9][10] we determine the product of effective \( g \)-factor and cyclotron mass, \( m^* g^* \), from the angular dependence of the SdH amplitudes and we find that \( g^* \) is enhanced compared to the free electron value.

We have fabricated field-effect transistors from SLG and BLG, by micromechanically exfoliating graphene flakes from graphite. The flakes were deposited on top of a Si/SiO₂ wafer, structured into a Hall-bar and covered with Au/Ti contacts [11]. Charge carriers are introduced by applying a gate voltage on the conducting Si substrate.

We present a detailed analysis on the spin splitting in a SLG sample made from Kish graphite with a mobility \( \mu = 0.8 \text{ Vm}^{-2}\text{s}^{-1} \) and BLG sample originating from natural graphite with a mobility \( \mu = 0.3 \text{ Vm}^{-2}\text{s}^{-1} \). Two other devices, one SLG and one BLG sample, showed qualitatively similar results.

To determine the spin-splitting we have measured the longitudinal resistances \( R_{xx} \) as a function of charge carrier concentration \( n \) at a constant perpendicular magnetic field. We adjusted the total magnetic field \( B_{tot} \) for each tilt angle such that the normal component \( B_n \) is the same (inset to Fig.1). The value of \( B_n \) was verified by measuring the Hall resistance of the devices in the non-quantized regime.

In Fig.1 we show the experimental \( R_{xx} \) dependencies for SLG at \( B_n = 6 \) T (a) and for BLG at \( B_n = 8 \) T (b). \( R_{xx} \) shows Shubnikov-de Haas oscillations with maxima whenever the Fermi energy is situated in the middle of a spin- and valley-degenerated Landau level \( E_N \), \( N = 0, 1, 2, ... \) being the Landau-level index. For the higher Landau levels \( \nu = 2 \) the longitudinal resistances do not exhibit zero minima indicating that the level broadening is comparable to the cyclotron energy at these perpendicular magnetic fields.

When increasing \( B_{tot} \) at a constant \( B_n \) the oscillation amplitudes for both BLG and SLG are reduced. From this reduction we determined the spin-splitting. We use the Lifshitz-Kosevich formula for systems with a general dispersion and we specifically include spin-splitting [9][10] with an effective \( g \)-factor \( g^* \) [12][13] and tilted magnetic fields [14]. The oscillatory contribution to the longitudinal resistance can be described as [2]:

\[
R_{xx} = A \cos \left( \frac{\hbar}{e B_n} S(E)|_{E=E_F} + \pi + \varphi_B \right)
\]  

where \( S(E)|_{E=E_F} \) is a extremal cross section of the Landau orbits in the \( k \)-space, \( A \) is the oscillation amplitude and \( \varphi_B \) is Berry phase, \( \varphi_B = \pi \) for SLG [1, 2].
\( \frac{A(0)}{A_0} \) and Eq. (1) yields the concentration-dependent resistance is given by \([12, 14]\):

\[
A = A_0(N) \cos \left( \frac{\pi g^* m^* B_{tot}}{2 m_e} \frac{B_{tot}}{B_n} \right) \tag{2}
\]

with cyclotron mass \([1]\):

\[
m^* = \frac{\hbar^2}{2\pi} \left. \frac{dS(E)}{dE} \right|_{E=E_F} \tag{3}
\]

and \(A_0(N)\) is constant for a given \(N\).

For the spherical Fermi surface in SLG and BLG with a Fermi wave-vector \(k_F = \sqrt{2m^*E_F}\), the extremal cross section of the Landau orbits is \(S(E)|_{E=E_F} = \frac{\pi k_F^2}{\hbar} = \pi n^2\) and Eq. (1) yields the concentration-dependent resistance oscillations as we observe them in our experiments:

\[
\tilde{R}_{xx} = A \cos \left( \frac{\hbar \pi^2}{e B_n} n + \pi + \varphi_B \right) = A \cos \left( \frac{\pi}{2} \nu + \pi + \varphi_B \right), \tag{4}
\]

\(\varphi_B = 2\pi\) for BLG \([3]\). The amplitude \(A\) contains a monotonic \(n\)-dependent part, a temperature dependence, a \(B_n\)-dependent contribution and a damping factor due to spin splitting depending on the total field \(B_{tot}\). At a constant temperature and perpendicular magnetic field this \(B_{tot}\)-dependence of the SdH amplitude \(A\) for charge carriers with cyclotron mass \(m^*\) and effective \(g\)-factor \(g^*\) is given by \([12, 14]\):

\[
A = A_0(N) \cos \left( \frac{\pi g^* m^* B_{tot}}{2 m_e} \frac{B_{tot}}{B_n} \right) \tag{2}
\]

To accurately determine the experimental oscillation amplitudes we have fitted our experimental data \(R_{xx}(n)\) to Eq. (4) in two steps. First we determined the oscillation period and a smooth background using all oscillations measured for a wide range of the carrier concentrations. Second we fitted the oscillation amplitudes \(A\) for each individual oscillation using the above determined period and background as fixed parameters. In Fig. 2 we show the final results of this fitting procedure for the SdH amplitude as a function of the total magnetic field for different Landau levels \(N\). For clarity all amplitudes are normalized to \(A_0\).

The experimentally observed reduction of the SdH amplitudes can be qualitatively visualized in a simple density of states (DOS) picture of a Landau level as depicted in Fig. 3a). In a purely perpendicular magnetic field the Landau level width exceeds the spin splitting and the DOS of the spin-down state (orange, horizontally dashed in Fig. 3a) overlaps with the one of the spin-up states (red, vertically dashed) to one broad Landau level. When increasing \(B_{tot}\) by leaving \(B_n\) constant, these two states move apart yielding an additional broadening of the Landau level with a reduced DOS in the center (green, solid areas in Fig. 3a). Eventually, when the spin splitting exceeds the level width a minimum between two distinct levels starts to develop in the DOS. This scenario is indeed observed experimentally in SLG (Fig. 3b). The SdH maxima corresponding to the \(N = 9\) and \(N = 10\) Landau levels in Fig. 3b) are fitted using Eq. (4) with a reduced DOS for the center area.

where \(\nu = (h n)/(e B_n)\) is the filling factor. As expected, the oscillation period, \((2eB_n)/(|\hbar\pi|)\), is independent on the band structure of the 2D material and only depends on the filling factor.
dual levels at $B_{tot} = B_n = 5$ T do not show any splitting. Increasing of the total field at a constant perpendicular component leads to a reduction of the oscillation amplitude and eventually appearance of spin-resolved peaks at the highest field of 28 T. However, this splitting is not yet enough to determine the energy difference by e.g. activation measurements.

A quantitative analysis of this decrease of the SdH amplitudes with increasing total magnetic field is done by fitting the data to Eq. 2 with $m^*g^*$ as a fitting parameter (solid lines in Fig. 2). The values for $m^*g^*$ obtained are plotted as a function of the charge carrier concentration in Fig. 3 for SLG (a) and BLG (b).

For both SLG and BLG the product $m^*g^*$ increases with concentration, which can be mainly attributed to the concentration dependent cyclotron mass $m^*$ of particles with a linear [1] and hyperbolic dispersion [15] as predicted by Eq. 3.

The dashed lines in Fig. 3a show the calculated dependence of $m^*g^*$ for $g^* = 2$ and $g^* = 2.7$ using $m^*(n) = (h/c) \sqrt{\pi n} [1]$. The shadowed areas represent a 10% uncertainty of this calculation mainly due to the experimental errors and some uncertainty in the Fermi velocity [10].

For SLG (Fig. 3a), the increase of $m^*g^*$ with $n$ is symmetric for electrons and holes (i.e. negative and positive $n$ in the figure). A best fit using $m^*(n)$ for SLG yields $g^* = 2.7 \pm 0.2$ (the error is the standard deviation). This finding is shown directly in the inset of Fig. 3a, where we plot the value of $g^*$ determined in the middle of each Landau level $N$ for different perpendicular fields $B_n$. Within an experimental error $g^*$ does not show any dependence on $N$ or $B_n$.

For BLG (Fig. 3b) the experimental situation is more complex as the observed increase of $m^*g^*$ with $n$ is not symmetric for holes and electrons. Such a behavior is caused by an asymmetry of $m^*$ resulting from an asymmetric band structure of biased BLG, which was already observed experimentally in transport experiments [17], cyclotron resonance [18] and activation-gap measurements [5]. Applying the experimental cyclotron mass from Ref. [17] (depicted as blue crosses in Fig. 4) allows us to estimate $g^*$ to be about 2.5 for both electrons and holes which is, within experimental accuracy, reasonably consistent with the g-factor enhancement observed in SLG.

The observed enhancement of the effective spin-splitting compared to its free-electron value can be explained by electron-electron interaction [19] yielding an interaction-enhanced splitting between two spin levels within one Landau level [20 [21].
\[ g^* \mu_B B_{tot} = g \mu_B B_{tot} + E_{ex}^0 (n_{\downarrow} - n_{\uparrow}) . \]  

(5)

Here \( g = 2 \) is a free-electron \( g \)-factor, \( E_{ex}^0 \) is an exchange parameter, and \( n_{\downarrow} \) and \( n_{\uparrow} \) are the relative occupations of the two spin states of a given Landau level.

For Gaussian shaped Landau levels with broadening \( \Gamma > g^* \mu_B B_{tot} \), i.e. where the spin splitting is not yet resolved, this relative occupation difference can be approximated using the Taylor expansion of the Gauss error function \( \text{erf}(g^* \mu_B B_{tot}/\Gamma) \):

\[ n_{\downarrow} - n_{\uparrow} \approx \sqrt{\frac{1}{2\pi}} \frac{g^* \mu_B B_{tot}}{\Gamma} \]  

(6)

and Eq. (5) yields:

\[ \frac{g^*}{g} = \left(1 - \frac{1}{\sqrt{2\pi}} \frac{E_{ex}^0}{\Gamma}\right)^{-1} . \]  

(7)

\( E_{ex}^0 \) is of the order of Coulomb interaction, \( E_{ex}^0 \propto \sqrt{\beta_0} \) [21], and \( \Gamma \propto \sqrt{B_\mu} \) [22]. Therefore, the ratio \( E_{ex}^0 / \Gamma \) remains constant and the \( g \)-factor enhancement is indeed predicted to be constant as we observe experimentally. Using the experimentally found \( g^* = 2.7 \) in Eq. (7) yields \( E_{ex}^0 = 130 \) K at 10 T when assuming \( \Gamma = 200 \) K [1][5]. For a completely spin polarized system, i.e. \( n_{\downarrow} - n_{\uparrow} = 1 \), one might then speculate that the exchange enhancement in the Eq. (5) would be an order of magnitude larger than a single particle Zeeman energy at this particular field.

Finally, we note, that the experimentally found enhanced values of \( g^* \) in graphene are close to those observed in transport experiments in graphite [23]. This may suggest that an exchange induced enhancement of \( g^* \) is quite common for graphitic materials. In contrast, no interaction-induced \( g \)-factor enhancement is observed using electron-spin resonance in graphene [24] and graphite [25] since these measurements are not sensitive to many body corrections [26]. Interestingly, measuring the Zeeman splitting of single-electron states in quantum dots, where no exchange enhancement of the \( g \)-factor is expected, also yields \( g \approx 2 \) [27], albeit with a considerable experimental uncertainty.

To conclude, we have experimentally measured and analyzed spin-splitting in SLG and BLG. We have shown that the product between the cyclotron mass \( m^* \) and the effective \( g \)-factor \( g^* \) increases with charge carrier concentration, as expected for a linear dispersion in SLG and a hyperbolic dispersion in BLG. Using the known concentration dependence of \( m^* \) we found that \( g^* \) in graphene is enhanced compared to the free-electron value and we attribute this to electron-electron interaction effects.

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