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.

The high 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/SiO$_2$ 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 $\mu = 0.8$ Vm$^{-2}$s$^{-1}$ and BLG sample originating from natural graphite with a mobility $\mu = 0.3$ Vm$^{-2}$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 $R_{xx}(n)$ 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 ($N \geq 2$) 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]:

$$R_{xx} = A \cos \left( \frac{\hbar}{eB_n} S(E)_{E=E_F} + \pi + \varphi_B \right)$$

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].
\( \varphi_B = 2\pi \) 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 \([12, 14]\):

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

with cyclotron mass \([1]\):

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

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

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

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

FIG. 1: (color online) Shubnikov de Haas oscillations in SLG (a) at \( T = 1.3 \) K and in BLG(b) at \( T = 0.4 \) K as a function of the carrier concentration for different total fields \( B_{tot} \) or tilt angles \( \theta \), respectively. When varying \( \theta \) the total field \( B_{tot} \) is adjusted such that the perpendicular field \( B_n \) remains constant, i.e. \( B_{tot} = B_n / \cos \theta \). The oscillation maxima are marked with the corresponding Landau level numbers \( N \). The inset schematically shows this tilting configuration.

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density of states for a Landau level with an increasing total magnetic field $B_{\text{tot}}$ (from the bottom to the top) at a constant perpendicular component $B_n$. Panel (b) shows this scenario as measured experimentally for the $N = 9, 10$ maximum in SLG at a constant perpendicular magnetic field $B_n = 5$ T.

Increasing of the total field at a constant perpendicular field $B_n$ does not show any splitting. The observed enhancement of the effective spin-splitting compared to its free electron mass $m_e$, as a function of charge carrier concentration for SLG (a) and BLG (b). The individual data points were extracted from the total-field dependence of the SdH amplitudes corresponding to different Landau levels $N = 2, ..., 10$ and represent measurement for a constant magnetic field $B_n = 5$ T, 6 T and 7 T for SLG and $B_n = 8$ T for BLG. The error bars represent the standard least squares fitting errors, taking into account error bars of $A$ (Fig. 2). The blue crosses in (a) represent the calculated behavior of $m^* g^*$ for different values of $g^*$ taking into account a 10% experimental uncertainty (shadowed areas). The blue crosses in (b) compare our data to the experimental cyclotron mass for BLG [17] multiplied by $g^* = 2.5$. The inset shows the effective $g$-factor, extracted from the product $m^* g^*$ in the main panel and the known cyclotron mass $m^*$ in SLG, as a function of Landau level index $N$.

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 $m^*(n) = (\hbar / c) \sqrt{\pi n}$. 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].
\[ g^* \mu_B B_{tot} = g \mu_B B_{tot} + E_{ex}^0 (n_\downarrow - n_\uparrow). \]  

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 \( \text{erf}(g^* \mu_B B_{tot}/\Gamma) \):

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

and Eq. (5) yields:

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

\( E_{ex}^0 \) is of the order of Coulomb interaction, \( E_{ex}^0 \propto \sqrt{N_n} \) [21], and \( \Gamma \propto \sqrt{B_n} \) [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 \( E_{ex}^0 \approx 130 \text{ K} \) at 10 T when assuming \( \Gamma = 200 \text{ K} \) [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 [21] 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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