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 (SdH) 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]:

$$\tilde{R}_{xx} = A \cos\left(\frac{\hbar}{eB_n} S(E)|_{E=E_F} + \pi + \varphi_B\right) \qquad (1)$$

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 φ_B is Berry phase, $\varphi_B = \pi$ for SLG [1, 2],

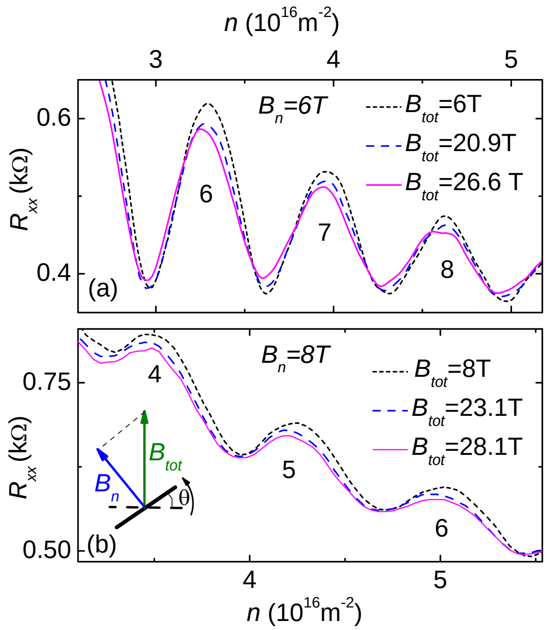

FIG. 1: (color online) Shubnikov de Haas oscillations in SLG (a) at $T = 1.3$ K and in BLG(b) at $T = 0.4$ K as a function of the carrier concentration for different total fields B_{tot} or tilt angles θ, respectively. When varying θ the total field B_{tot} is adjusted such that the perpendicular field 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.

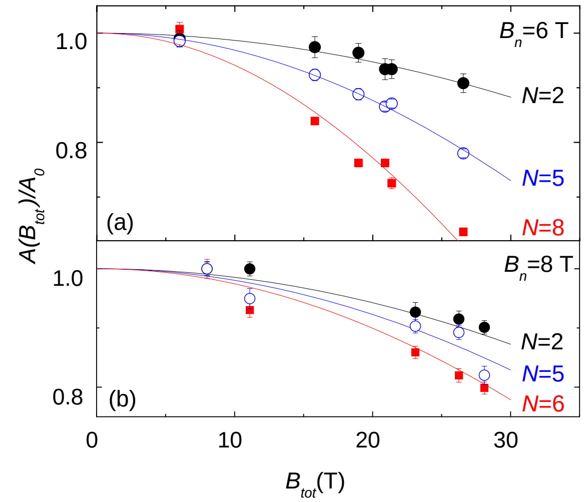

FIG. 2: (color online) Normalized oscillation amplitudes as a function of total field B_{tot} at a constant perpendicular field B_n in SLG (a) and BLG (b). Error bars represent standard least squares fitting errors in the determination of A. Solid lines are fits to Eq. 2 with m^*g^* as a fit parameter.

$\varphi_B = 2\pi$ 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]:

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

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:

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

where $\nu = (hn)/(eB_n)$ 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.

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 $N = 10$ Lan-

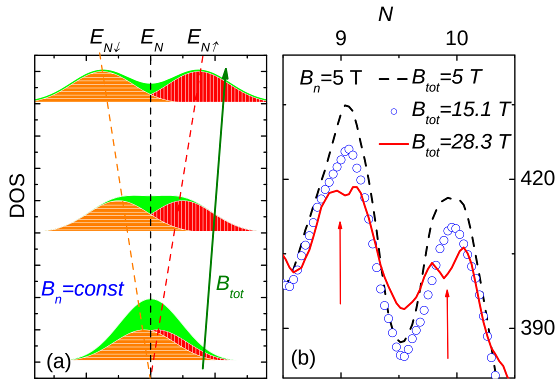

FIG. 3: (color online) Schematic representation of the density of states for a Landau level with an increasing total magnetic field B_{tot} (from the bottom to the top) at a constant perpendicular component B_n (a). Panel (b) shows this scenario as measured experimentally for the $N = 9, 10$ maximum in SLG at a constant perpendicular magnetic field $B_n = 5$ T.

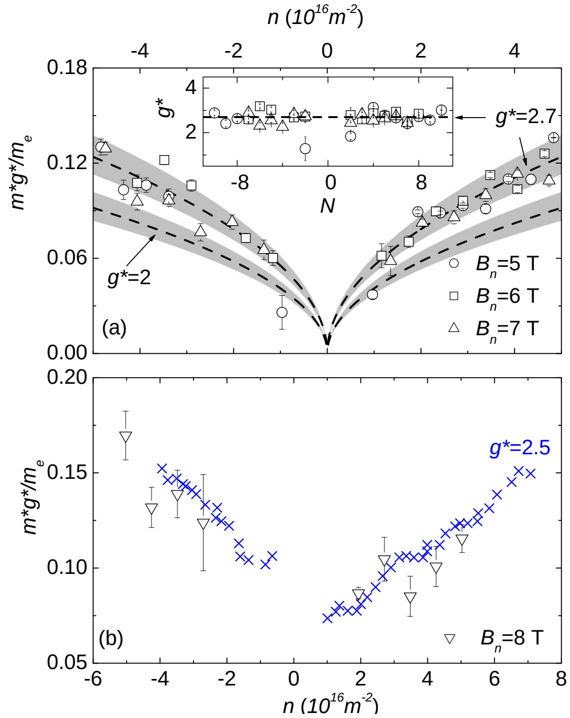

FIG. 4: (color online) Experimentally deduced m^*g^* (open symbols), normalized to the free electron mass m_e, as a function of charge carrier concentration for SLG (a) and BLG (b). The individual data points were extracted from the total-field dependence of the SdH amplitudes corresponding to different 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 dashed lines 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.

dau 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. 4 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. 4a show the calculated dependence of m^*g^* for $g^* = 2$ and $g^* = 2.7$ using $m^*(n) = (\hbar/c)\sqrt{\pi n}$ [1]. The shadowed areas represent a 10% uncertainty of this calculation mainly due to the experimental errors and some uncertainty in the Fermi velocity [16].

For SLG (Fig. 4a), 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. 4a, 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. 4b) 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]:

$$g^*\mu_B B_{tot} = g\mu_B B_{tot} + E^0_{ex}(n_\downarrow - n_\uparrow). \qquad (5)$$

Here $g = 2$ is a free-electron g-factor, E^0_{ex} is an exchange parameter, and $n_\uparrow$ and $n_\downarrow$ 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 $\mathrm{erf}(g^*\mu_B B_{tot}/\Gamma)$:

$$n_\downarrow - n_\uparrow \approx \sqrt{\frac{1}{2\pi}}\,\frac{g^*\mu_B B_{tot}}{\Gamma} \qquad (6)$$

and Eq.(5) yields:

$$\frac{g^*}{g} = \left(1 - \sqrt{\frac{1}{2\pi}}\frac{E^0_{ex}}{\Gamma}\right)^{-1}. \qquad (7)$$

E^0_{ex} is of the order of Coulomb interaction, $E^0_{ex} \propto \sqrt{B_n}$ [21], and $\Gamma \propto \sqrt{B_n}$ [22]. Therefore, the ratio E^0_{ex}/Γ 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 $E^0_{ex} = 130$ K at 10 T when assuming $\Gamma = 200$ K [4, 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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