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. By 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. By using the cyclotron mass for a single-layer and a bilayer graphene, we find an enhanced g factor \(g^*\) = 2.7 ± 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 \(B\rangle\) = 20 T,4 which allows an experimental adjustment the total magnetic field \(B\rangle\) = 20 T.7 Another observation of a spin degeneracy lifting within one Landau level is negligible smaller compared to the normal component \(B\rangle\) = 20 T.7.8

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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 \(B\rangle\) for each tilt angle such that the normal component \(B_n\) is the same (see the inset to Fig. 1). The value of \(B_n\) was verified by measuring the Hall resistance of the devices in the nonquantized 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 \(E_N\), \(N = 0, 1, 2, \ldots\) being the Landau-level index. For the higher Landau levels (\(N > 2\)) 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 \(B\rangle\) 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 \(g^*\) (Refs. 12 and 13) and tilted magnetic fields.14

The oscillatory contribution to the longitudinal resistance can be described as:

\[
R_{xx} = A \cos \left( \frac{\hbar}{eB_n} S(E)_{E=E_f} + \pi + \phi_B \right),
\]

where \(S(E)_{E=E_f}\) is an extremal cross section of the Landau orbits in the k space, \(A\) is the oscillation amplitude, and \(\phi_B\) is the Berry phase, \(\phi_B = \pi\) for SLG,1,2 and \(\phi_B = 2\pi\) 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 \(B\rangle\). At a constant temperature and perpendicular magnetic field this \(B\rangle\) dependence of the ShD amplitude \(A\) for charge carriers with cyclotron mass \(m^*\) and effective g factor \(g^*\) is given by:

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

where \(m_e\) is the electron mass, \(B_{tot}\) is the total magnetic field, and \(A_0(n)\) is a function of the charge-carrier concentration.
such that the perpendicular field $B_n$ and Landau-level numbers $B_n/\cos \theta$ are normalized to using the above determined period and background as fixed magnetic field for different Landau levels.

To accurately determine the experimental oscillation amplitudes as a function of the total field $B_{\text{tot}}$ or tilt angles $\theta$, respectively. When varying $\theta$, the total field $B_{\text{tot}}$ is adjusted such that the perpendicular field $B_n$ remains constant, i.e., $B_{\text{tot}} = B_n/\cos \theta$. The oscillation maxima are marked with the corresponding Landau-level numbers $N$. The inset schematically shows this tilting configuration.

with cyclotron mass

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

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:

$$\tilde{R}_{xx} = A \cos \left( \frac{\hbar^2}{2\pi} n + \pi + \psi_B \right) = A \cos \left( \frac{\pi}{2} \nu + \pi + \psi_B \right).$$

where $\nu = (\hbar n)/(e B_n)$ is the filling factor. As expected, the oscillation period $(2e B_n)/(\hbar \pi)$ is independent on the band structure of the two-dimensional 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 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. 3(a). 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. 3(a)] overlaps with the one of the spin-up states (red, vertically dashed) to one broad Landau level. When increasing $B_{\text{tot}}$ 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. 3(a)]. 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. 3(b)]. The SdH maxima corresponding to the $N = 9$ and $N = 10$ Landau levels at $B_{\text{tot}} = B_n = 5$ T do not show any splitting. Increasing

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig1.png}
\caption{(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 $B_{\text{tot}}$ or tilt angles $\theta$.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig2.png}
\caption{(Color online) Normalized oscillation amplitudes as a function of total field $B_{\text{tot}}$ 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.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig3.png}
\caption{(Color online) Schematic representation of the 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 $B_n$ (a). (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.}
\end{figure}
the total field at a constant perpendicular component leads to a reduction of the oscillation amplitude and eventually to the 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 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 and hyperbolic dispersion as predicted by Eq. (3).

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The dashed lines in Fig. 4(a) show the calculated dependence of $m^* g^*$ for $g^* = 2$ and $g^* = 2.7$ using $m^* = (\hbar/e c)^2 \nabla n$. The shaded areas represent a 10% uncertainty of this calculation, mainly due to the experimental errors and some uncertainty in the Fermi velocity. For SLG [Fig. 4(a)], 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 $g^* = 2.7 \pm 0.2$ (error is the standard deviation). This finding is shown directly in the inset of Fig. 4(a), 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. 4(b)] 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, cyclotron resonance, and activation-gap measurements. Applying the experimental cyclotron mass from Ref. 17 (depicted as crosses in Fig. 4) allows us to estimate $g^*$ to be $\sim 2.5$ 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 an electron-electron interaction yielding an interaction-enhanced splitting between two spin levels within one Landau level.4-6

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

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 by using the Taylor expansion of the Gauss error function erf($g^* \mu_B B_{tot}/\Gamma$):

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

and Eq. (5) yields

$$\frac{g^*}{g} = \left(1 - \sqrt{\frac{1}{2\pi \Gamma}} E_{ex}^0 \frac{1}{\Gamma}ight)^{-1}.$$
transport experiments in graphite. 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 and graphite since these measurements are not sensitive to many-body corrections. Interestingly, measuring the Zeeman splitting of single-electron states in quantum dots, where no exchange enhancement of the \( g \)-factor is expected, also yields \( g \approx 2 \), 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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