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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 $g^* = 2.7 \pm 0.2$ 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 Rapid Communication 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 the 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 $\mu = 0.8$ V m⁻² s⁻¹ and a BLG sample originating from natural graphite with a mobility $\mu = 0.3$ V m⁻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 (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 , $N = 0, 1, 2, \dots$ 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^* (Refs. 12 and 13) and tilted magnetic fields.¹⁴ The oscillatory contribution to the longitudinal resistance can be described as²

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

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 φ_B is the Berry phase, $\varphi_B = \pi$ for SLG,^{1,2} and $\varphi_B = 2\pi$ 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}

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

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., $B_{\text{tot}} = B_n / \cos \theta$. The oscillation maxima are marked with the corresponding Landau-level numbers N . The inset schematically shows this tilting configuration.

with cyclotron mass¹

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

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:

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

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

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.

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_{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

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). (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.

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 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 linear¹ and hyperbolic dispersion¹⁵ as predicted by Eq. (3).

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 $N = 2, \dots, 10$ and represent measurements for a constant magnetic field $B_n = 5, 6$, and 7 T for SLG and $B_n = 8 \text{ T}$ for BLG. The error bars represent the standard least-squares-fitting errors, taking into account the 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 crosses in (b) compare our data to the experimental cyclotron mass for BLG (Ref. 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 .

The dashed lines in Fig. 4(a) 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. 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 $g^* = 2.7 \pm 0.2$ (the 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,¹⁷ cyclotron resonance,¹⁸ and activation-gap measurements.⁵ Applying the experimental cyclotron mass from Ref. 17 (depicted as crosses in Fig. 4) allows us to estimate g^* to be ~ 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 interaction¹⁹ yielding an interaction-enhanced splitting between two spin levels within one Landau level:^{20,21}

$$g^* \mu_B B_{\text{tot}} = g \mu_B B_{\text{tot}} + E_{\text{ex}}^0 (n_{\downarrow} - n_{\uparrow}). \quad (5)$$

Here $g = 2$ is a free-electron g factor, E_{ex}^0 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 $\Gamma > g^* \mu_B B_{\text{tot}}$, 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 $\text{erf}(g^* \mu_B B_{\text{tot}} / \Gamma)$:

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

and Eq. (5) yields

$$\frac{g^*}{g} = \left(1 - \sqrt{\frac{1}{2\pi}} \frac{E_{\text{ex}}^0}{\Gamma} \right)^{-1}. \quad (7)$$

E_{ex}^0 is of the order of the Coulomb interaction $E_{\text{ex}}^0 \propto \sqrt{B_n}$,²¹ and $\Gamma \propto \sqrt{B_n}$.²² Therefore, the ratio E_{ex}^0 / 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 $E_{\text{ex}}^0 = 130 \text{ K}$ at 10 T when assuming $\Gamma = 200 \text{ K}$.^{4,5} For a completely spin-polarized system, i.e., $n_{\downarrow} - n_{\uparrow} = 1$, one might then speculate that the exchange enhancement in 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 graphene²⁴ and graphite²⁵ 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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