

Quantum Hall States near the Charge-Neutral Dirac Point in Graphene

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We investigate the quantum Hall (QH) states near the charge-neutral Dirac point of a high mobility graphene sample in high magnetic fields. We find that the QH states at filling factors $\nu = \pm 1$ depend only on the perpendicular component of the field with respect to the graphene plane, indicating that they are not spin related. A nonlinear magnetic field dependence of the activation energy gap at filling factor $\nu = 1$ suggests a many-body origin. We therefore propose that the $\nu = 0$ and ± 1 states arise from the lifting of the spin and sublattice degeneracy of the $n = 0$ Landau level, respectively.

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The experimental observation of the quantum Hall (QH) effect in single atomic sheet of graphene [1,2] has attracted much attention recently, particularly due to the unique electronic transport observed in this material. Compared with the conventional integer QH effect in many other two-dimensional (2D) systems, the Hall resistance (R_{xy}) quantization condition in graphene is shifted by a half-integer: $R_{xy}^{-1} = \pm g_s(n + 1/2)\frac{e^2}{h}$, where n is the Landau level (LL) index, e is the electron charge, h is Planck's constant, and $g_s = 4$ is the LL degeneracy, accounting for spin and sublattice symmetry in graphene. This quantization condition leads to the QH effect appearing at filling factors $\nu = \pm 2, \pm 6, \pm 10, \dots$. It is now understood that this unique QH effect is related to the quasirelativistic nature of the charge carriers in graphene [3–5], stemming from the unusual linear dispersion relation of its bands near the charge-neutral Dirac point in the graphene band structure [6,7].

More recently, the QH effect in graphene has been studied in the extreme quantum limit in a very strong magnetic field [8]. New QH states, corresponding to filling factors $\nu = 0, \pm 1, \pm 4$, are clearly resolved in magnetic fields $B > 20$ T, indicating a lifting of the fourfold degeneracy of the $n = 0$ LL and a twofold degeneracy of the $n = \pm 1$ LLs, respectively. While angular dependent activation energy gap measurements indicate that the QH states at $\nu = \pm 4$ are spin states, the origin of the QH states at $\nu = 0, \pm 1$ remains unresolved.

The nature of these QH states near the charge-neutral Dirac point is of fundamental interest. There have been numerous theoretical investigations [9–25] of these states, and their origin is currently under considerable debate. Recently, Abanin *et al.* [26] suggested that the $\nu = 0$ QH state is spin-polarized and dissipative, owing to counter-propagating edge states at the charge-neutral point, supported by a finite metallic resistivity at low temperatures.

In this Letter, we present an experimental investigation of the QH states near the Dirac point. We find that the $\nu =$

± 1 states depend only on the out of plane component of the applied magnetic field, and show a rather large energy gap with an approximately square root dependence on the magnetic field. This suggests a many-particle origin of this splitting as it would originate from the breaking of the sublattice degeneracy of the $n = 0$ LL at the Dirac point. As a consequence, and by elimination, our results would imply that the $\nu = 0$ QH gap is induced by the lifting of the spin degeneracy.

Our sample is a high-quality graphene specimen with mobility as high as $\sim 2 \times 10^4$ cm²/V s measured at carrier density $n_e = 4 \times 10^{12}$ cm⁻². The graphene sheet is mechanically extracted from Kish graphite following a method similar to the one described in [1,2]. The sample is deposited onto a Si substrate, which serves as a gate electrode separated from the sample by 300 nm of insulating SiO₂. To perform transport measurement, multiple electrodes are patterned in van der Pauw geometry (inset to Fig. 1) using conventional electron beam lithography, followed by Au/Cr (30/3 nm) thermal evaporation and a standard lift-off process. The electronic transport is measured over the temperature range of 4.2–300 K, using a lock-in technique. The sample is mounted on a single-axis tilting stage to allow *in situ* tuning of the angle, $\theta = \cos^{-1}(B_p/B_{\text{tot}})$, where B_{tot} is the total magnetic field and B_p is the component perpendicular to the graphene plane.

The fourfold degeneracy of the $n = 0$ LL of graphene consists of a twofold degeneracy from the spin symmetry and a twofold degeneracy from the sublattice symmetry. One may be able to distinguish the origin of any particular splitting by performing magnetotransport measurement in a tilted field, where a spin splitting depends on B_{tot} , whereas a sublattice splitting (caused by electron-electron correlations) would only depend on B_p . Figure 1 shows the measured magnetoresistance R_{xx} with respect to the back gate voltage V_g at a temperature of $T = 4.2$ K and in two different magnetic fields: $B_{\text{tot}} = 45$ T, $B_p = 20$ T (dashed

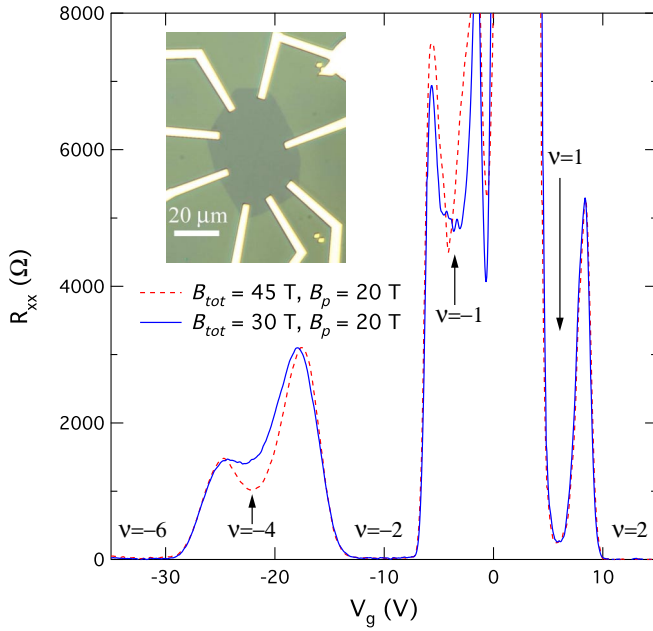


FIG. 1 (color online). Magnetoresistance R_{xx} with respect to the back gate voltage V_g at a temperature of $T = 4.2$ K and in two different magnetic fields: $B_{\text{tot}} = 45$ T, $B_p = 20$ T (dashed curve); and $B_{\text{tot}} = 30$ T, $B_p = 20$ T (solid curve). The minimum magnetoresistance R_{xx}^{min} substantially increases as B_{tot} decreases for $\nu = -4$ state, while for the $\nu = \pm 1$ states R_{xx}^{min} practically does not depend on B_{tot} for the same B_p .

curve); and $B_{\text{tot}} = 30$ T, $B_p = 20$ T (solid curve). Note that in this comparison we use the same B_p but different B_{tot} . The minimum magnetoresistance R_{xx}^{min} substantially increases as B_{tot} decreases for $\nu = -4$ state, while for the $\nu = \pm 1$ states R_{xx}^{min} hardly varies with B_{tot} for the same B_p . This observation supports the spin related origin of the $\nu = \pm 4$ splittings [8], and also, more importantly, suggests that the $\nu = \pm 1$ states are likely due to the breaking of the orbital degeneracy of the sublattice symmetry in the $n = 0$ LL.

In order to further characterize the nature of $\nu = \pm 1$ QH states, we measure the activation energy of R_{xx}^{min} at fixed magnetic fields. Figure 2(b) displays the Arrhenius plots of R_{xx}^{min} of the $\nu = 1$ state [27]. A well-defined thermal activation behavior is readily observable ($R_{xx}^{\text{min}} \sim \exp[-\Delta E/2k_B T]$, where k_B is the Boltzmann constant), and the corresponding energy gap, ΔE , can be extracted for different magnetic fields. In Fig. 2(a), we plot the obtained energy gaps at $\nu = 1$, denoted as $\Delta E(\nu = 1)$, as a function of B field. For comparison, we have also reproduced the measured $\Delta E(\nu = \pm 4)$ of the same sample from Ref. [8]. We find $\Delta E(\nu = 1)$ to be considerably larger than $\Delta E(\nu = \pm 4)$. For instance, at 45 T, $\Delta E(\nu = 1) \geq 4\Delta E(\nu = \pm 4)$. Moreover, unlike for $\Delta E(\nu = \pm 4)$, which showed a linear B -field dependence, the B -field dependence of $\Delta E(\nu = 1)$ does not seem to follow such a simple relationship. Forcing a linear fit onto $\Delta E(\nu = 1)$ produces a positive y intercept

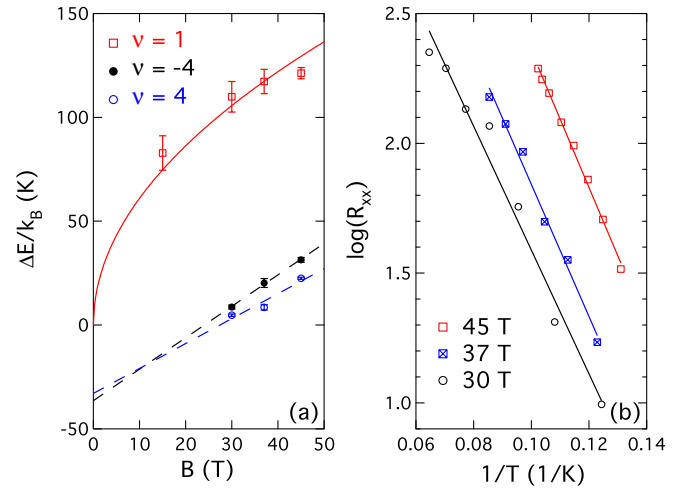


FIG. 2 (color online). (a) The measured activation energy gap ΔE as a function of magnetic field for the QH states at filling factors $\nu = 1$ (square), $\nu = -4$ (solid dot), and $\nu = 4$ (open dot). While $\Delta E(\nu = \pm 4)$ are linear with respect to B field (data are reproduced from Ref. [8]), $\Delta E(\nu = 1)$ can be fitted better by a \sqrt{B} dependence (solid curve). (b) Arrhenius plots of R_{xx} minimum of the $\nu = 1$ state for three different magnetic fields: 45, 37, and 30 T. The straight lines are linear fits to the data.

which would indicate an unphysical, negative LL energy width. In fact, a \sqrt{B} behavior provides a better fit to the $\Delta E(\nu = 1)$ data as shown in the solid curve in Fig. 2(a).

The lack of a linear dependence and the existence of a roughly \sqrt{B} dependence point to a nonspin origin and possibly to a many-particle origin of the gap, and suggest that the $\nu = \pm 1$ states are associated with a spontaneous breaking of the sublattice symmetry driven by the electron-electron interactions [13–21]. In this picture, $\Delta E(\nu = \pm 1)$ is expected to be on the scale of $e^2/\epsilon l_B$ [16], and thus proportional to \sqrt{B} , where $l_B = \sqrt{\hbar/eB}$ is the magnetic length and ϵ is the dielectric constant. We calculate that $e^2/\epsilon l_B \sim 1100$ K, assuming $\epsilon = 4$ and $B = 45$ T. This value is much larger than the Zeeman energy $E_Z = g\mu_B B \sim 60$ K at $B = 45$ T, where $g = 2$ is the g factor and μ_B is the Bohr magneton. This simple evaluation indicates that $\Delta E(\nu = \pm 1) \gg E_Z$ in the experimentally accessible magnetic field, suggesting the importance of electron-electron interaction under magnetic fields.

In Fig. 3, we summarize our current understanding of the sequence of the QH states near the charge-neutral Dirac point of graphene in a schematic of the LL hierarchy. We use up arrows and down arrows to represent the spin of the charge carriers, and solid (blue) and open (red) dots for different valleys in the graphene band structure. Since our measurements suggest that the $\nu = \pm 1$ states are associated with the valley splitting of the $n = 0$ LL due to electron-electron correlations, the QH state at $\nu = 0$ must be related to the spin splitting of this LL. However, we also note that the behavior of R_{xx} and R_{xy} at $\nu = 0$ is completely different from that of any other QH states away

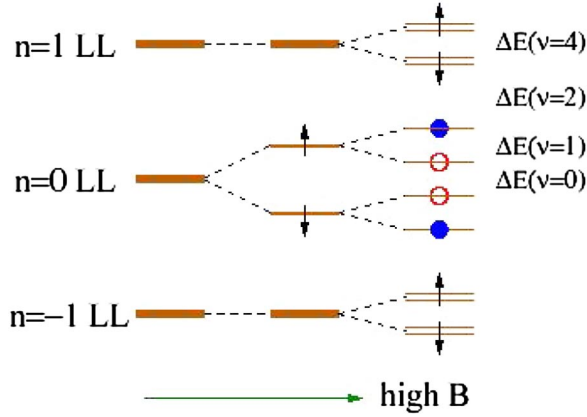


FIG. 3 (color online). Schematic of the LL hierarchy in graphene in magnetic fields. The up and down arrows represent the spin of the charge carriers, and the solid (blue) and open (red) dots indicate different valleys in the graphene electronic band.

from the charge-neutral Dirac point. Unlike usual QH states, the $\nu = 0$ QH state does not show a resistance minimum in R_{xx} nor a clear resistance plateau in R_{xy} . This state only becomes visible as a plateau in the Hall conductance. Figure 4 displays R_{xx} vs V_g near the Dirac point over a wide range of temperatures. No activation behavior has been observed at $\nu = 0$. Recently, Abanin *et al.* provided a possible interpretation for the existence of this state as the consequence of countercirculating edge states with opposite spin [26]. Such a state would be consistent with our proposal shown in Fig. 3.

We now address the relative size of the energy gap between the levels displayed in Fig. 3. In a sufficiently

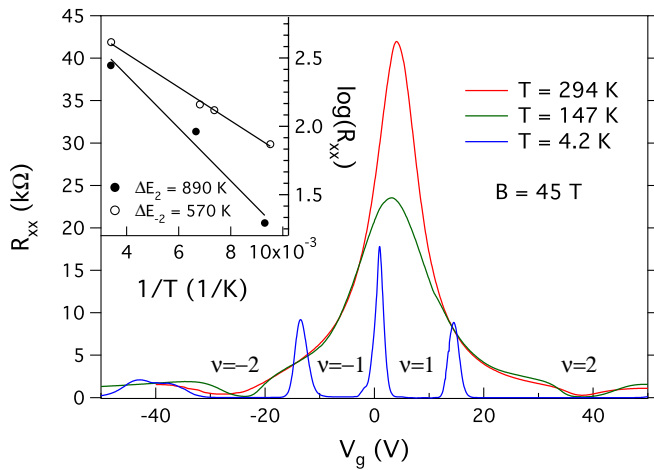


FIG. 4 (color online). Magnetoresistance R_{xx} as a function of the back gate voltage V_g at $B = 45$ T in a wide temperature range, from room temperature to liquid helium temperature. A robust QH effect is clearly seen even at room temperature. No activated behavior has been observed in R_{xx} of the $\nu = 0$ state (at Dirac point). Inset: Arrhenius plots of R_{xx} minimum of the $\nu = \pm 2$ states at $B = 45$ T. The straight lines are linear fits to the data.

large magnetic field, typically $B > 20$ T, the QH states in high-quality graphene specimens are robust, even at room temperature [28] (see also Fig. 4). From the inset to Fig. 4, we estimate the activation energy gaps of the QH states at $\nu = \pm 2$ and $B = 45$ T to be $\Delta E(\nu = 2) \approx 890$ K (solid dots), and $\Delta E(\nu = -2) \approx 570$ K (open dots). In a simplistic view, we may interpret these values as the gap between the $n = 0$ and the $n = \pm 1$ LLs in Fig. 3. In a single-particle picture with unlifted spin and sublattice degeneracies [29], the LL energy spectrum in graphene can be described by

$$E_n = \text{sgn}(n)\sqrt{2\hbar v_F^2 B |n|}, \quad (1)$$

where v_F is the Fermi velocity of graphene with a typical value of $v_F \approx 10^6$ m/s [1,2,30,31]. Hence, at $B = 45$ T, the calculated energy spacing between the $n = 0$ and the $n = 1$ LL would be $E_{0 \rightarrow 1} \approx 2800$ K. This value is more than 3 times larger than the largest measured energy gap $\Delta E(\nu = 2) \approx 890$ K. We believe that such a large discrepancy cannot be accounted for in terms of spin or sublattice symmetry splitting, nor by any reasonable LL broadening. We first rule out the possibility of an enhanced spin splitting since the g factor is not enhanced by exchange in a completely filled LL. With the bare value of $g = 2$, the spin splitting of the LL reaches only $g\mu_B B \sim 60$ K at 45 T. We further eliminate the sublattice degeneracy splitting as a potential explanation of the observed discrepancy, as this gap $\Delta E(\nu = \pm 1)$ must collapse when the Fermi energy lies between the $n = 0$ and the $n = 1$ LL. Finally, the LL broadening due to scattering may lead to a reduction of the energy gap. However, we infer such a reduction is negligible compared to $E_{0 \rightarrow 1} \approx 2800$ K: from our low temperature measurements ($T = 1.4$ K) [8], we estimate a LL broadening of $\Gamma \approx 20$ K. Since the mobility of graphene changes only by $\sim 30\%$ from 30 mK to room temperature [32], Γ is likely irrelevant on the scale of $E_{0 \rightarrow 1}$.

At this stage we do not know what causes this strong reduction of the measured LL energy gap as compared to the calculated one. One may speculate that many-particle effects are partially responsible for the discrepancy. Recent infrared experiments in graphene [33–35] between LL levels $n = 0$ and $n = \pm 1$ yield a rather good agreement with Eq. (1). Yet theory suggests that as much as 30% of this energy is due to many-particle corrections [36]. Since both single-particle energy level and many-particle correction have \sqrt{B} -field dependences in graphene, they cannot be separated. This would suggest that the Fermi velocity (typically $v_F \approx 10^6$ m/s) can be as much as $\sim 30\%$ smaller in reality, the difference being made up for by many-particle corrections. If activation energy measurements were much less affected by such corrections, the expected gaps would follow the bare value of v_F , leading to a $\sim 30\%$ reduction from Eq. (1). This would bring the calculated energy gap of $E_{0 \rightarrow 1} \approx 1900$ K closer to the measured gap of $\Delta E(\nu = 2) \approx 890$ K, but still leaves a

substantial, unresolved discrepancy. The implication of such a reinterpretation of ν_F would be considerable and more extensive studies of the LL spectrum in graphene will be required to verify such a trend.

In conclusion, we study the QH states in graphene at filling factors $\nu = \pm 1$ in tilted magnetic fields and elevated temperatures. Our results indicate that the $\nu = \pm 1$ QH states originate from the lifting of the sublattice symmetry of the $n = 0$ LL caused by electron-electron interactions. Measurements of the activation energy gaps of the QH states near the Dirac point indicate a significant deviation from a simplistic single-particle model, which suggests that many-particle effects need to be taken into account to understand the LLs near the charge-neutral Dirac point.

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