The Nature of 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 them to be not spin-related. A non-linear 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 $\pm 1$ states arise from the lifting of the spin and sub-lattice degeneracy of the $n = 0$ LL, 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: $R_{xy}^\pm = \pm g_s (n + 1/2) e^2 / h$, 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 sub-lattice symmetry in graphene. This quantization condition leads to the QH effect appearing at filling factors $\nu = \pm 2, \pm 6, \pm 10, \ldots$. It is now understood that this unique QH effect is related to the quasi-relativistic nature of the charge carriers in graphene [3,4,5], stemming from the unusual linear dispersion reflecting the unique electronic transport observed in this material [3,4].

More recently, the QH effect in graphene has been studied in the extreme quantum limit in a very strong magnetic field [6]. 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 four-fold degeneracy of the $n = 0$ LL and a two-fold degeneracy of the $n = \pm 1$ LLs respectively. While angular dependent activation energy gap measurements indicate that the QH states at $\nu = \pm 1$ 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 [1,4,10,11,12] of these states, and their origin is currently under considerable debate. Recently, Abanin et al. [24] 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 $\nu = \pm 1$ 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 sub-lattice 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 $\sim 2 \times 10^4$ cm$^2$/Vs 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 SiO$_2$. 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, $\theta = \cos^{-1}(B_p / B_{tot})$, where $B_{tot}$ is the total magnetic field and $B_p$ is the component perpendicular to the graphene plane.

The four-fold degeneracy of the $n = 0$ LL of graphene consists of a two-fold degeneracy from the spin symmetry and a two-fold degeneracy from the sub-lattice 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 sub-lattice splitting (caused by electron-electron correlations) would only depend on $B_p$. Figure 1 shows the measured magnetoresistance, $R_{xx}$, with re-
spect to the back gate voltage, $V_g$, at a temperature of $T = 4.2$ K and in two different magnetic fields: $B_{tot} = 45$ T, $B_p = 20$ T (dashed curve); and $B_{tot} = 30$ T, $B_p = 20$ T (solid curve). The minimum magnetoresistance $R_{xx}^{\text{min}}$ substantially increases as $B_{tot}$ decreases for $\nu = -4$ state, while for the $\nu = \pm 1$ states $R_{xx}^{\text{min}}$ practically does not depend on $B_{tot}$ for the same $B_p$.

In order to further characterize the nature of $\nu = \pm 1$ QH states, we measure the activation energy of $R_{xx}^{\text{min}}$ at fixed magnetic fields. Figure 2(b) displays the Arrhenius plots of $R_{xx}^{\text{min}}$ of the $\nu = 1$ state. A well-defined thermal activation behavior is readily observable ($R_{xx}^{\text{min}} \sim \exp[-\Delta E/2k_BT]$, where $k_B$ is the Boltzmann constant), and the corresponding energy gap, $\Delta E$, 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 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 non-spin 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 sub-lattice symmetry driven by the electron-electron interactions [13, 14, 15, 16, 17, 18, 19, 20, 21]. In this picture, $\Delta E(\nu = \pm 1)$ is expected to be on the scale of $e^2/\epsilon l_B \approx 1100$ 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 $\mu_B$ is the Bohr magneton. This simple evaluation indicates that $\Delta E(\nu = \pm 1) >> E_Z$ 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 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 counter-circulating edge states with opposite spin. 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 large magnetic field, typically \( B > 20 \) T, the QH states in high-quality graphene specimens are robust, even at room temperature. 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 un-lifted spin and sub-lattice degeneracies, the LL energy spectrum in graphene can be described by

\[ E_n = \text{sgn}(n) \sqrt{2e\hbar v_F^2 B |n|}, \tag{1} \]

where \( v_F \) is the Fermi velocity of graphene with a typical value of \( v_F \approx 10^6 \text{ m/s} \). Hence, at \( B = 45 \) T, the calculated energy spacing between the \( n = 0 \) and the \( n = 1 \) LL would be \( E_{0-1} \approx 2800 \) K. This value is more than three 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 sub-lattice 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 sub-lattice 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 \( E_{0-1} \approx 2800 \) 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 \[32\], \( \Gamma \) is likely irrelevant on the scale of \( E_{0-1} \).

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 \[33, 34\] 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 \[32\]. Since both single-particle energy level and many-particle correction have \( \sqrt{B} \)-field dependences in graphene, they cannot be separated. This would sug-
gest that the Fermi velocity (typically $v_F \approx 10^6 \text{ m/s}$) 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 $E_{0-1} \approx 1900 \text{ K}$ closer to the measured gap of $\Delta E(\nu = 2) \approx 890 \text{ K}$, but still leaves a substantial, unresolved discrepancy. The implication of such a reinterpretation of $v_F$ 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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