Quantum Hall effect in graphene

Z. Jiang\textsuperscript{a,b}, Y. Zhang\textsuperscript{a}, Y.-W. Tan\textsuperscript{a}, H.L. Stormer\textsuperscript{a,c}, P. Kim\textsuperscript{a,}\textsuperscript{∗}

\textsuperscript{a} Department of Physics, Columbia University, New York, NY 10027, USA
\textsuperscript{b} National High Magnetic Field Laboratory, Tallahassee, FL 32310, USA
\textsuperscript{c} Department of Applied Physics and Applied Mathematics, Columbia University, New York, NY 10027, USA

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Abstract

The quantum Hall (QH) effect in two-dimensional electron and hole gas is studied in high quality graphene samples. Graphene samples whose lateral size $\sim 10 \mu m$ were fabricated into mesoscopic devices for electrical transport measurement in magnetic fields. In an intermediate field range of up to 10 T, a distinctive half-integer QH effect is discovered with QH plateaus appearing at a filling factor sequence, $\nu = 4(n + 1/2)$, where $n$ is the Landau level (LL) index. As the magnetic field increases to the extreme quantum limit, we observe additional QH plateaus at filling factors $\nu = 0, \pm 1, \pm 4$. Further detailed investigations show that the presence of the $\nu = 0, \pm 1$ QH states indicates the $n = 0$ LL at the charge neutral Dirac point splits into four sublevels. This lifts both the sublattice and the spin degeneracy, while the QH states at $\nu = \pm 4$ can be attributed to lifting of the spin degeneracy of the LLs. Above 30 T of magnetic field, the large quasiparticle gaps between the $n = 0$ and $n = \pm 1$ LLs lead to the QH effect that can be observed even at room temperature.

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1. Introduction

Over the last twenty years there has been a tremendous interest in the electronic properties of low dimensional condensed matter systems (e.g. two-dimensional (2D) electrons/holes confined in semiconductor devices; mesoscopic heterostructures incorporating thin metallic, superconducting, and/or ferromagnetic films). The interest in these systems mainly stems from the fact that, as device dimensions approach fundamental microscopic length scales, novel quantum effects manifest themselves. The quantum electronic properties of such systems are commonly described by the Schrödinger equation, in which quasiparticles behave non-relativistically with a finite effective mass. Recently, great interest has arisen in another class of condensed matter system, where the charge carrier dynamics appears effectively relativistic with zero effective carrier mass and the transport properties are governed by the Dirac equation [1–3]. This fascinating and unique system is graphene: a single atomic sheet of graphitic carbon atoms that are arranged into a honeycomb lattice.

The hexagonal arrangement of carbon atoms in graphene can be decomposed into two interpenetrating sublattices of carbon atoms with inversion symmetry between them. This unique topology provides an unusual energy dispersion relation [5]. The energy dispersion near the charge neutrality points, termed “Dirac points”, is of particular interest, since there the 2D energy spectrum is linear, and thus the electrons always move at a constant speed, the band velocity $v_F \approx 10^6$ m/s. Many of the interesting electronic properties in graphene result from this dispersion relation which is analogous to that of relativistic, massless fermions. In fact, even before the experimental discovery of graphene, the distinct nature of charge carriers in a hexagonal carbon lattice has been speculated upon and was proposed to have important implications on the electronic transport in graphitic materials, including single-walled carbon nanotubes [6,7].
A sudden burst of experimental and theoretical work on graphene was initiated by the recent work that demonstrated experimentally accessible single and multi layered graphene samples by mechanical extraction [8–11,13] and chemical synthesis [12]. Ever since, numerous unique electrical, chemical, and mechanical properties of graphene have been discovered. In particular, a very unusual half-integer quantum Hall effect (QHE) and a non-zero Berry’s phase [14,15] were discovered in graphene, which provide the unambiguous evidence of the existence of Dirac fermions in graphene and distinguish it from other conventional 2D electronic systems with a finite carrier mass.

In this paper, we review the QHE which appears in graphene based on the recent published work [14–16]. We first describe the sample fabrication and characterization method based on mechanical extraction. Then we discuss the experimental results on the QHE in high mobility graphene samples in the intermediate field regime ($B < 10$ T) followed by the QHE in the extreme quantum limit ($B > 20$ T).

2. Sample preparation

The experimental discovery of an unusual half-integer QHE in graphene was possible because of the significant progress in device fabrication techniques for single layer graphene samples [8,13], and in particular the success in obtaining high mobility in such devices. The high mobility graphene samples used in our experiments were extracted from Kish graphite (Toshiba Ceramics Co.) or other natural graphite single crystals. Single layers of graphene were deposited on silicon wafers, following a method similar to the one described in Ref. [13]. This method involves a very simple, yet efficient method for mechanical extraction of mesoscopic graphite pieces. First, single crystals of graphite are fixed on an adhesive tape and cleaved to expose a fresh surface. Next, very thin flakes of graphite pieces are transferred from the crystals to the surface of silicon wafer by bringing the cleaved surface of the single crystals in contact with the wafer. Various thicknesses are deposited on the surface of SiO$_2$/Si substrate including single layers of graphite with a lateral size 1–100 μm. Optical microscope images show color contrast in samples of different thickness due to interference induced color shifts. These color shifts are sensitive enough to determine the number of graphene layers on top of the SiO$_2$/Si substrate. We also note that the thickness of SiO$_2$ layer affects the contrast between different layers. 300 nm thick thermally grown SiO$_2$ layer yields the best contrast for this purpose.

After identifying suitable samples, we choose graphene pieces of lateral size 3–10 μm for device fabrication. Multiple electrodes, arranged in Hall bar (Fig. 1(a)) or van der Pauw geometry (Fig. 1(b)), are fabricated on the sample using electron beam lithography followed by Cr/Au (3/30 nm) evaporation and a lift-off process. For samples with non-ideal geometry, or for smaller single-layer pieces connected to larger pieces of thicker graphite, a second electron beam lithography step was performed to define an etch mask on the top of graphene. We then use an oxygen plasma etching process to remove unnecessary parts of the samples (Fig. 1(c)).

For the longitudinal and Hall resistance measurement, we apply a small AC current ($I < 10$ nA) through the samples and measure the transverse voltage ($V_{xy}$) and longitudinal voltage ($V_{xx}$) using a lock-in amplifier. For the Hall bar geometry, the magnetoresistance $R_{xx}$ and Hall resistance $R_{xy}$ are then obtained from $R_{xx} = V_{xx}/I$ and $R_{xy} = V_{xy}/I$, respectively (Fig. 1(d)). The degenerately doped silicon substrate serves as a gate electrode with silicon oxide acting as the gate dielectric. By applying a gate bias voltage, $V_g$, the charge density of the sample can be tuned. The induced charge density can be derived from a Hall measurement, where a magnetic field $B$ is applied perpendicular to the sample surface, and a linear increase $R_{xy}$ ($B$) is observed. In this measurement, we obtain the carrier density $n_e$ from the slope of these lines: $n_e = 1/(e(dR_{xy}/dB))$. The mobility of the sample is given by $\mu = (L/W)(e n_e R_{xx})^{-1}$, where $W$ and $L$ are the width and length of the Hall bar. The gate voltage corresponding to the charge neutrality point (i.e., $n_e = 0$) is often found shifted to $V_{Dirac}$, indicating a natural charge doping from the environment to the samples.

Although $\mu$ defined in this way diverges near $n_e \approx 0$ due to the failure of the semiclassical Drude model near the Dirac points of graphene, the limiting value of the mobility, $\mu_L$, defined in the large density limit ($n_e \approx 5 \times 10^{12}$ cm$^{-2}$) serves as a good indicator of the sample quality. The majority of samples we measured so far exhibit $\mu_L$ values in the 2000–20,000 cm$^2$/V s range. Empirically, we found that the samples with $\mu_L > 5000$ cm$^2$/V s show the QHE in the intermediate field regime ($4 < B < 10$ T). Only the highest quality samples, with quality ($\mu_L > 15,000$ cm$^2$/V s), exhibit structures beyond half-integer QHE in the extreme quantum limit ($B > 20$ T).

3. Half-integer quantum Hall effect in graphene

Exceptionally high mobility graphene samples allow us to investigate transport phenomena in the magnetic quantum limit...
where the QHE manifests itself. Fig. 2 shows $R_{xy}$ and $R_{xx}$ of a typical high mobility ($\mu > 10,000 \text{ cm}^2/\text{V s}$) graphene sample as a function of magnetic field $B$ at a fixed gate voltage $V_g > V_{\text{Dirac}}$. The overall positive $R_{xy}$ indicates that the contribution is mainly from electrons. At high magnetic field, $R_{xy}(B)$ exhibits plateaux and $R_{xx}$ is vanishing, which are the hallmark of the QHE. At least two well-defined plateaux with values $(2e^2/h)^{-1}$ and $(6e^2/h)^{-1}$, followed by a developing $(10e^2/h)^{-1}$ plateau, are observed before the QHE features transform into Shubnikov–de Haas (SdH) oscillations at lower magnetic field. We observed the equivalent QHE features for holes $V_g < V_{\text{Dirac}}$ with negative $R_{xy}(B)$ values.

Alternatively we can access the QH plateaux by tuning the electron density by adjusting $V_g$ at a fixed magnetic field. Fig. 3 shows $R_{xy}$ of the sample of Fig. 2 as a function of gate voltage $V_g$ at $B = 9 \text{ T}$. A series of fully developed QH states, i.e., plateaux in $h/(e^2\nu)$ quantized to values with an integer filling factor $\nu$, are observed, which are the hallmark of the QHE. Well-defined $\nu = \pm 2, \pm 6, \pm 10, \pm 14$ QH states are clearly seen, with quantization according to

$$R_{xy}^{-1} = \pm 4 \left( n + \frac{1}{2} \right) \frac{e^2}{h}$$

where $n$ is a non-negative integer, and $+/-$ stands for electrons and holes respectively. This quantization condition can be translated into the quantized filling factor $\nu = \pm 4(n + 1/2)$ in the usual QHE language. While the QHE has been observed in many 2D systems, the QHE observed in graphene is distinctively different from those 'conventional' QHE's since the quantization condition Eq. (1) is shifted by a half integer.

This so-called half-integer QHE is unique to graphene. It has been predicted by several theories which combine ‘relativistic’ Landau levels (LLs) with the particle–hole symmetry of graphene [1–3]. The experimental phenomena can be understood from the calculated LL spectrum in the Dirac spectrum.

As in other 2D systems, application of a magnetic field $B$ normal to the graphene plane quantizes the in-plane motion of charge carriers into LLs. The LL formation for electrons/holes in graphene has been studied theoretically using an analogy to $2+1$ dimensional Quantum Electro Dynamics (QED) [4], in which the LL energy is given by

$$E_n = \text{sgn}(n) \sqrt{2\hbar v_F^2 |n| B}.$$  \hspace{1cm} (2)

Here $e$ and $\hbar$ are electron charge and Planck’s constant divided by $2\pi$, and the integer $n$ represents an electron-like ($n > 0$) or a hole-like ($n < 0$) LL index. In particular, a single LL with $n = 0$ also occurs, where electrons and holes are degenerate. Note that in Eq. (2), we do not consider a spin degree of freedom, assuming the separation of $E_n$ is much larger than the Zeeman spin splitting. Therefore each LL has a degeneracy $g_s = 4$, accounting for spin degeneracy and sublattice degeneracy. This assumption needs to be changed when the magnetic field becomes large as we will discuss in the next section.

The observed QH sequence can be understood employing the symmetry argument for the Hall conductivity $\sigma_{xy} = -R_{xy}/(R_{xy}^2 + (W/L)^2 R_{xx}^2)$, where $L$ and $W$ are the length and width of the sample, respectively. With the given LL spectrum in Eq. (2), the corresponding Hall conductance $\sigma_{xy}$ exhibits QH plateaux when an integer of LLs are fully occupied, and jumps by an amount of $g_s e^2/h$ when the Fermi energy, $E_F$, crosses a LL. Time reversal invariance guarantees particle–hole symmetry and thus $\sigma_{xy}$ is an odd function in energy across the Dirac point [4]. Here, in particular, the $n = 0$ LL is pinned at zero energy. Thus the first plateau for electrons ($n = 1$) and holes ($n = -1$) are situated exactly at

![Fig. 2. Quantized magnetoresistance and Hall resistance of a graphene device where $n \approx 10^{12} \text{ cm}^{-2}$ and $T = 1.6 \text{ K}$. The horizontal lines correspond to the inverse of the multiples $e^2/h$. The QHE in the electron gas is demonstrated by at least two quantized plateaux in $R_{xy}$ with vanishing $R_{xx}$ in the corresponding magnetic field regime.](image_url)

![Fig. 3. The Hall resistance as a function of gate voltage at fixed magnetic field $B = 9 \text{ T}$, measured at 1.6 K. The horizontal lines correspond to the inverse of integer multiples of $e^2/h$ values.](image_url)
As the magnetic field grows, the LL spectrum assumed in Eq. (2) is no longer valid due to increasing Zeeman spin splitting energy and many-body related correlation effect in the extreme quantum limit. Fig. 4 shows experimentally obtained $\sigma_{xy}$ increases (decreases) by an amount of $g_s e^2/h$, which yields the quantization condition in Eq. (1).

4. Graphene quantum Hall effect in high magnetic fields

As the magnetic field grows, the LL spectrum assumed in Eq. (2) is no longer valid due to increasing Zeeman spin splitting energy and many-body related correlation effect in the extreme quantum limit. Fig. 4 shows experimentally obtained $\sigma_{xy}$ at $B = 25$ T as a function of $V_g$. A series of fully developed QH states with plateaus quantized to values $g_s(n + 1/2)e^2/h$ are observed. These well-defined QH states in $\sigma_{xy}$ are in accordance with the previous low-magnetic field measurements ($B < 20$ T) shown in Fig. 3. In addition to these QH states, new QH states at $\nu = 0$ and at $\nu = \pm 1, \pm 4$ are clearly resolved in the high magnetic field data $B > 20$ T. These new QH states can be attributed to the magnetic field induced splitting of the $n = 0$ and $n = \pm 1$ LLs [16]. In lower magnetic fields, each LL at energy $E_n$ is assumed to be four-fold degenerate due to a two-fold spin degeneracy and a two-fold sublattice symmetry. The observed filling factor sequence $\nu = 0$ and $\pm 1$ in higher fields ($B > 20$ T) implies that these degeneracies of $n = 0$ LL are fully lifted, such that $\sigma_{xy}$ increases in steps of $e^2/h$ as $E_F$ passes through the LLs. On the other hand, the four-fold degeneracy of the $n = \pm 1$ LLs is only partially resolved, leaving a two-fold degeneracy in each of these LLs and $\sigma_{xy}$ increases in steps of $2e^2/h$.

The broken sublattice symmetry in graphene in high magnetic fields is quite surprising, because it has been known that the presence of a magnetic field alone does not break the inversion symmetry of the graphene lattice. Since the sublattice symmetry is protected by the inversion symmetry of graphene, single-electron type of perturbations such as simple uniaxial strain does not lift the graphene sublattice degeneracy. This points to many-body electron correlations within LLs as an alternative origin for the lifting of the degeneracy at the Dirac point. There have been numerous theoretical discussion for the possible origin of this observed degeneracy lifting in the $n = 0$ LL [17–31].

The QH plateau located right at the Dirac point ($V_g \approx V_{\text{Dirac}}$) is particularly interesting. While $\sigma_{xy}$ exhibits a clearly resolved $\nu = 0$ plateau, the observed $R_{xx}$ always shows a finite peak rather than zero for $\nu = 0$ state. Thus this new type of QH state does not conform to the standard QH observation. Recent studies focused on this state reveals [32,33] that the $\nu = 0$ QH state results from the splitting of the spin degeneracy, leaving two coexisting edge states circulating in opposite direction with opposite spin [32].

In order to further investigate the origin of the $\nu = \pm 4$ QH states, we have carried out detailed temperature dependence measurements to determine the energy gaps between neighboring LLs, $\Delta E$ [16]. In particular, we measured the temperature dependence of the associated $R_{xx}$ minimum, $R_{xx}^{\text{min}}$, at $\nu = \pm 4$. The inset of Fig. 5 shows the Arrhenius plots which reveal thermally activated behavior, $R_{xx}^{\text{min}} \propto \exp[-\Delta E/(2k_B T)]$, where $k_B$ is the Boltzmann constant. We find that $\Delta E$, derived from linear fits to these data, depends linearly on the magnitude of total magnetic field $B_{\text{tot}}$ (the main section of Fig. 5). Our experimental results suggest that the energy gap of the QH states can be written as [34]: $\Delta E = g^* \mu_B B_{\text{tot}} - 2\Gamma$, where $g^*$ is the effective $g$ factor, $\mu_B$ is the Bohr magneton, and $\Gamma$ is the half-width of the LL broadening at half-maximum. We assume that $\Gamma$ is constant, then $g^*$ and $\Gamma$ can be extracted from the slope and y-intercept of the linear fit and find $g^* = 2.0$ and $\Gamma = 18.2$ K from Fig. 5 for the QH state at $\nu = -4$ and $g^* = 1.7$ and $\Gamma = 15.8$ K for the $\nu = +4$ QH state from a
Fig. 6. The magnetoresistance and Hall resistance as a function of gate voltage at fixed magnetic field $B = 45$ T, measured at 300 K. The horizontal lines correspond to the inverse of the multiples of $e^2/h$ values.

5. Summary

Mesoscopic graphene devices are fabricated employing high mobility graphene samples mechanically extracted from graphite single crystals. Magnetoresistance and Hall resistance measurements show the unusual half-integer quantum Hall effect in graphene. We find that the quantization condition in graphene is distinctively different from conventional 2D systems by a shift of a half-integer. In the extreme quantum limit, we observe that $n = 0$ LL splits into four sublevels, lifting both spin and sublattice degeneracy while $n = \pm 1$ LLs lift the spin degeneracy only. Detailed temperature dependence measurements show that the effective $g$ factor at filling factors $\nu = \pm 4$ is close to the bare electron $g$ factor. Due to the large quasiparticle gaps between the $n = 0$ and $n = \pm 1$ LLs, a robust QHE can be observed at room temperature.

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