Unconventional Integer Quantum Hall Effect in Graphene

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Monolayer graphite films, or graphene, have quasiparticle excitations that can be described by (2 + 1)-dimensional Dirac theory. We demonstrate that this produces an unconventional form of the quantized Hall conductivity $\sigma_{xy} = -(2e^2/h)(2n + 1)$ with $n = 0, 1, \ldots$, which notably distinguishes graphene from other materials where the integer quantum Hall effect was observed. This unconventional quantization is caused by the quantum anomaly of the $n = 0$ Landau level and was discovered in recent experiments on ultrathin graphite films.

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The quantum Hall effect (QHE) is one of the most remarkable phenomena in condensed matter discovered in the second half of the 20th century. The basic experimental fact characterizing QHE is that the diagonal electric conductivity of a two-dimensional electron system in a strong magnetic field is vanishingly small $\sigma_{xx} \to 0$, while the nondiagonal conductivity is quantized in multiples of e^2/h: $\sigma_{xy} = -\nu e^2/h$, where ν is an integer [the integer quantum Hall effect (IQHE)] or a fractional number (the fractional QHE). In a recent paper [1] the fabrication of freestanding monocrystalline graphite films with thickness down to a single atomic layer was reported. This new material, called graphene, possesses truly remarkable properties such as excellent mechanical characteristics, scalability to the nanometer sizes, and the ability to sustain huge ($> 10^8$ A/cm^2) electric currents. By using the electric-field effect [1], it is possible to change the carrier concentration in samples by ten times and even to change the carrier type from electron to hole when the sign of applied gate voltage is altered. All this makes graphene a promising candidate for applications in future microelectronics and nanoelectronics.

On the theoretical side, the linear, Dirac-like, spectrum of quasiparticle excitations (up to energies of the order of 1000 K) and the pseudospin degeneracy make graphene a unique truly two-dimensional "relativistic" electronic system. The thinnest graphite films can be described by a low-energy (2 + 1)-dimensional effective *massless* Dirac theory [2]. Of special interest are the properties of graphene in a magnetic field. The important differences between the Dirac and Schrödinger theories may be observed in thermodynamic and magnetotransport measurements [1,3–6]. For instance, the phase of de Haas–van Alphen and Shubnikov–de Haas oscillations for Dirac quasiparticles is shifted [7–10] compared to the phase of nonrelativistic quasiparticles. Moreover, the Dingle and temperature factors in the amplitude of oscillations explicitly depend on the carrier density in the case of a Dirac-like spectrum [7,8].

Because of the large value of the cyclotron gap, it is expected that the QHE in this material can be observed for much higher temperatures and lower magnetic fields than in conventional semiconductors. Therefore it is natural to ask whether the fundamental difference between the properties of Landau levels (LL) [see Eqs. (5) and (10) below] in the Dirac and Schrödinger theories can be observed experimentally in the Hall conductivity. The purpose of this Letter is to show that the Dirac-like dynamics of graphene results in an unconventional form of the Hall quantization

$$\sigma_{xy} = -\frac{2e^2}{h}(2n + 1), \qquad n = 0, 1, \ldots. \tag{1}$$

We argue that the quantization rule (1) is caused by the *quantum anomaly of the $n = 0$ LL*, i.e., by the fact that it has *a twice smaller degeneracy* than the levels with $n > 0$ and its energy does not depend on the magnetic field [11]. Remarkably this quantization *is observed* experimentally [10] for ultrathin graphite films that exhibit the behavior expected for ideal 2D graphene.

We begin with the Lagrangian density of noninteracting quasiparticles in a single graphene sheet that in the continuum limit reads [2]

$$\mathcal{L} = \sum_{\sigma=\pm 1} \bar{\Psi}_\sigma \left[i\gamma^0(\hbar\partial_t - i\mu_\sigma) + iv_{\mathrm{F}}\gamma^i\left(\hbar\partial_i - i\frac{e}{c}A_i\right)\right]\Psi_\sigma, \tag{2}$$

where $\Psi_\sigma = (\psi_{1\sigma}(t, \mathbf{r}), \psi_{2\sigma}(t, \mathbf{r}))$ is the four-component Dirac spinor combined from two spinors $\psi_{1\sigma}$ and $\psi_{2\sigma}$ (corresponding to $\mathbf{K}$ and $\mathbf{K}'$ points of the Fermi surface, respectively) that describe the Bloch states residing on the two different sublattices of the biparticle hexagonal lattice of the graphene sheet, and $\sigma = \pm 1$ is the spin. In Eq. (2) γ^μ with $\mu = 0, 1, 2$ are 4×4 γ matrices belonging to a reducible representation in 2 + 1, $\bar{\Psi}_\sigma = \Psi^\dagger_\sigma\gamma_0$ is the Dirac conjugated spinor, $-e < 0$ is the electron charge, v_{F} is the Fermi velocity. We set $k_B = 1$, but kept Planck constant $\hbar = h/2\pi$.

The external magnetic field **B** is applied perpendicular to the plane along the positive z axis and the vector potential is taken in the symmetric gauge $\mathbf{A} = (-B/2y, B/2x)$. In contrast to the truly relativistic (3 + 1) case [12], the Zeeman interaction term still has to be explicitly added to the Lagrangian (2), because it originates from nonrelativistic many-body theory. This can be done by considering spin splitting $\mu_\sigma = \mu - \sigma\mu_B B$ of the chemical potential μ, where $\mu_B = e\hbar/(2mc)$ is the Bohr magneton. However, for realistic values of $v_F \propto 10^5$ m/s in graphene the distance between LL is very large compared to the Zeeman splitting [8], so that in what follows we do not consider this term and simply multiply all relevant expressions by 2 to count the spin degeneracy. While simple tight-binding calculations made for the hexagonal lattice of a single graphene sheet predict that $\mu = 0$, the real picture is more complicated and the actual value of μ can be nonzero due to finite doping and/or disorder. Moreover, nonzero and even tunable values of μ (including the change of the character of carriers, either electrons or holes) is possible in electric-field doping experiments [1,4,10]. In our notations $\mu > 0$ corresponds to electrons and accordingly to the positive gate voltage.

Using the Kubo formalism and modeling the LL by Lorentzians with a constant width Γ, the following expression for the diagonal conductivity was obtained in Refs. [8,13]

$$\sigma_{xx}(B, \mu, \Gamma) = \frac{2e^2}{\hbar} \int_{-\infty}^{\infty} d\omega[-n_F'(\omega - \mu)]\mathcal{A}_{xx}(\omega, B, \Gamma), \tag{3}$$

where $n_F(\omega) = 1/[\exp(\omega/T) + 1]$ is the Fermi distribution and the function $\mathcal{A}_{xx}$ that incorporates the effect of all LL is given by Eq. (11) of Ref. [8]. Now this result is extended for the Hall conductivity, and we derive a general analytical expression for $\sigma_{xy}(B, \mu, \Gamma)$ [14]. The resulting dependence $\sigma_{xy}(\mu)$ is shown in Fig. 1, where one sees that the plateaux of σ_{xy} follow Eq. (1). This agrees with the latest experimental results [10] and resembles earlier theoretical predictions [15].

However, to demonstrate result (1) in the most transparent way, it is useful to write down a simpler conventional representation [13,16] for σ_{xy} obtained in the clean limit $\Gamma \to 0$:

$$\sigma_{xy} = -\frac{ec\rho}{B} \equiv -\frac{e^2\,\mathrm{sgn}(eB)\,\mathrm{sgn}\mu}{\pi\hbar}\,\nu_B. \tag{4}$$

Here we introduced the filling factor of LL, $\nu_B = \pi\hbar c|\rho|/|eB|$ with ρ being the carrier imbalance ($\rho \equiv n_e - n_h$, where n_e and n_h are the densities of "electrons" and "holes," respectively). This filling factor can be represented as a sum over LL

$$M_n = \sqrt{\Delta^2 + 2n\hbar v_F^2|eB|/c}, \qquad n = 0, 1, \ldots, \tag{5}$$

of the Dirac theory:

$$\mathrm{sgn}\,\mu\,\nu_B = \frac{1}{2}\Big[\tanh\frac{\mu + \Delta}{2T} + \tanh\frac{\mu - \Delta}{2T} + 2\sum_{n=1}^{\infty}\Big(\tanh\frac{\mu + M_n}{2T} + \tanh\frac{\mu - M_n}{2T}\Big)\Big], \tag{6}$$

where we separated out the level with $n = 0$ because its degeneracy is only half of the degeneracy of the levels with $n > 0$. To illustrate this rather peculiar property of the Dirac theory in a perspicuous way, we included in M_n and ν_B the mass (excitonic gap) Δ that was discussed recently to explain some experiments [13,17]. Our consideration of σ_{xy} is, in fact, independent of the presence of Δ, so in what follows we set $\Delta = 0$. A zero value of Δ is expected for noninteracting quasiparticles on the hexagonal lattice of graphene.

The first equality in Eq. (4) corresponds to a classical straight line $\sigma_{12} \propto \nu_B$. As discussed, for example, in Ref. [18], this line emerges from two step function dependences, viz. $\mu(\rho)$ and $\sigma_{12}(\mu)$. Indeed, using $\tanh(\omega/2T) = \mathrm{sgn}(\omega)$ for $T \to 0$, we obtain from

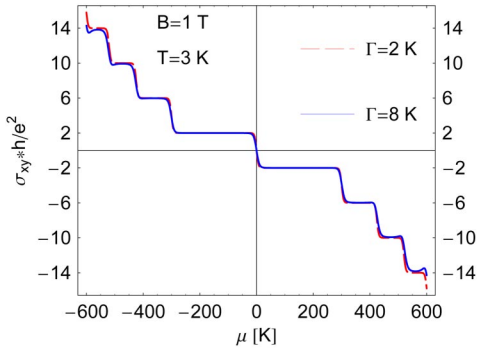

FIG. 1 (color online). The Hall conductivity σ_{xy} measured in e^2/h units as a function of chemical potential μ for two different values of Γ for $T = 3$ K and $B = 1$ T. We use $\hbar v_F^2 eB/c \to (4.5 \times 10^4\ \mathrm{K}^2)B(\mathrm{T})$.

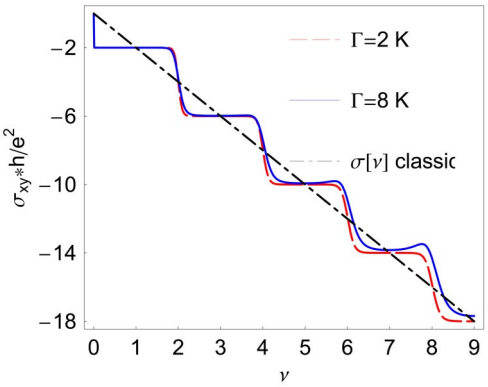

FIG. 2 (color online). The Hall conductivity σ_{xy} measured in e^2/h units as a function of the filling ν_B. The straight line corresponds to a classical dependence (4). The parameters are the same as in Fig. 1.

Eqs. (4) and (6) (compare with Ref. [16])

$$\sigma_{xy} = -\frac{2e^2\,\mathrm{sgn}(eB)\,\mathrm{sgn}\mu}{h}\left(1 + 2\left[\frac{\mu^2 c}{2\hbar|eB|v_F^2}\right]\right), \quad (7)$$

where $[x]$ denotes the integer part of x. The usual argumentation (see, e.g., Ref. [18]) for the occurrence of the IQHE states that in the presence of disorder the dependence of $\sigma_{12}(\mu)$ remains the same, while $\mu(\rho)$ becomes a smooth function. The classical (4) and quantum (7) Hall conductivities coincide only for the fillings, $\nu_B = 2n + 1$ (see Fig. 2).

The odd integer rather than integer fillings that produces the quantization rule (1) appears due to the above-mentioned halved degeneracy of the $n = 0$ LL. Another interesting feature of Eq. (7) (see also Figs. 1 and 2) is that $\sigma_{xy} = \pm 2e^2/h$ for the fillings $\nu_B < 1$, and it crosses 0 only when μ changes sign. On the contrary, in a conventional IQHE $\sigma_{xy} = 0$ for $\nu_B < 1$.

Although Eqs. (4)–(7) are obtained in the clean limit and using a simple bare bubble expression for conductivity, our main result (1) is model independent and is based only on the $n = 0$ level anomaly.

Now we rewrite Eqs. (4)–(6) in terms of the Fermi distribution

$$\sigma_{xy} = -\frac{2e^2\,\mathrm{sgn}(eB)}{h}\sum_{n=0}^{\infty}(2n+1)[n_F(M_n - \mu) + n_F(-M_n - \mu) - n_F(M_{n+1} - \mu) - n_F(-M_{n+1} - \mu)] \quad (8)$$

to compare it with Eq. (18) of Ref. [19] that was obtained for an ideal two-dimensional electron gas

$$\sigma_{xy} = -\frac{e^2}{h}\sum_{n=0}^{\infty}(n+1)[n_F(\omega_n^{\mathrm{nonrel}}) - n_F(\omega_{n+1}^{\mathrm{nonrel}})] \quad (9)$$

with nonrelativistic spectrum

$$\omega_n^{\mathrm{nonrel}} = \frac{e\hbar B}{mc}\left(n + \frac{1}{2}\right) - \mu. \quad (10)$$

There is a commonsense reasoning [1,20] that graphene is a two-band (the first band corresponds to the electrons with $\omega_n = M_n - \mu$ and the second band to the holes with $\omega_n = -M_n - \mu$), two-valley (corresponding to $\mathbf{K}$ and $\mathbf{K}'$ points of graphen's Fermi surface) semiconductor with zero gap Δ between the bands. Accordingly, its Hall conductivity can be directly obtained from (9) by summing over all these bands and valleys

$$\sigma_{xy}^{\mathrm{semicond}} = -\frac{2e^2\,\mathrm{sgn}(eB)}{h}\sum_{n=0}^{\infty}2(n+1)[n_F(M_n - \mu) + n_F(-M_n - \mu) - n_F(M_{n+1} - \mu) - n_F(-M_{n+1} - \mu)], \quad (11)$$

where we also counted spin degeneracy. It is easy to see that Eqs. (8) and (11) correspond to two completely different Hall conductivity quantization rules, viz. Eq. (8), which correctly counts the degeneracy of the $n = 0$ level, produces Eq. (1), while the semiconducting analogy (11) leads to

$$\sigma_{xy}^{\mathrm{semicond}} = -\frac{4e^2}{h}n, \qquad n = 0, 1, \ldots. \quad (12)$$

Here we assumed that $e, B, \mu > 0$. Although previous experimental observations supported the picture based on Eq. (12), the latest experiments made on thin films [10] are in accord with the unconventional Hall quantization (1). This shows that in an applied magnetic field the semiconducting interpretation of graphene's band structure that led us to Eq. (11) becomes invalid (see also Ref. [21]). The drastic difference between Eqs. (1) and (12) is caused by the above-mentioned fact that the lowest LL in Dirac theory is special and has twice smaller degeneracy than the levels with $n > 0$, because, depending on the sign of eB, it is occupied either by electrons or by holes, while higher levels contain both electrons and holes [11,22]. In the nonrelativistic theory when the Landé factor $g \neq 2$ all Landau levels have the same degeneracy [12]. It turns out that graphene, for which the valence and conduction bands intersect in discrete points [23], is reasonably well described by the Dirac formalism which naturally embodies the $n = 0$ level anomaly.

We now consider the phenomenon of quantum magnetic oscillations in graphene, which is closely related to the quantization of σ_{xy}, and discuss the specifics of the $n = 0$ level. The de Haas–van Alphen and Shubnikov–de Haas effects in graphene were studied in Refs. [7–9]. In particular, in Ref. [8] it was shown that the oscillatory part of the diagonal conductivity (3) is given by

$$\sigma_{xx} \propto \sum_{k=1}^{\infty}\cos\left[\frac{\pi k\mu^2}{\hbar v_F^2|eB|/c}\right]R_T(k)R_D(k)R_s(k), \quad (13)$$

where R_T, R_D, and R_s are, respectively, the temperature, Dingle, and spin factors. Using the relationship $\mu^2 = \pi\hbar^2 v_F^2|\rho|$ valid for $T = \Gamma = B = 0$ [13], one can check that the minima of the diagonal conductivity (3) occur at the fillings $\nu_B = 2n + 1$, giving an indication of the possible positions of the plateaux in the IQHE [10]. (Note that in thick films the minima of σ_{xx} occur at integer fillings [4].) Obviously for $\mu = 0$ there is no oscillations of σ_{xx}, the conductivity $\sigma_{xx}(\mu = 0) = 2e^2/(\pi^2\hbar)$ becomes a field independent universal [24] quantity that is another distinctive feature of the $n = 0$ level anomaly.

Although the quantization (1) can be understood by considering noninteracting Dirac quasiparticles placed in an external magnetic field, even this simple model reveals other unusual properties [11] intimately related to nontrivial dynamics of quasiparticles from the $n = 0$ level. For example, the $U(4)$ symmetry of the Lagrangian (2) is spontaneously broken down to $U(2) \times U(2)$ at $\mu = 0$ in nonzero magnetic field even in the absence of additional interaction between fermions [11], thus leading to the emergence of the chiral condensate $\langle\bar{\Psi}\Psi\rangle$. Including

many-body effects such as an attractive interaction between quasiparticles could further generate a gap for quasiparticles like the above-mentioned gap Δ (see, e.g., Refs. [13,17]). Fortunately in the case of the IQHE the presence of the condensate does not affect our consideration. On the other hand, a possible gap generation for the fermions from the lowest LL might become important for the fractional quantum Hall effect, and this issue certainly deserves further experimental and theoretical study.

To conclude, we have shown that the integer numbers associated with quantized Hall conductivity in graphene have an unusual pattern $\sigma_{xy}h/e^2 = 2, 6, 10, 14\ldots$. We argued that it is related to the fact that a theoretical description of graphene is based on $(2 + 1)$-dimensional Dirac theory, where the lowest Landau level has half of the higher Landau levels degeneracy.

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