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Electron–hole coexistence in disordered graphene probed by high-field magneto-transport

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Abstract. We report on magneto-transport measurement in disordered graphene under a pulsed magnetic field of up to 57 T. For large electron or hole doping, the system displays the expected anomalous integer quantum Hall effect (IQHE) specific to graphene down to the filling factor \( \nu = 2 \). In the close vicinity of the charge neutrality point, the system breaks up into coexisting puddles of holes and electrons, leading to a vanishing Hall and finite longitudinal resistance with no hint of divergence at very high magnetic field. Large resistance fluctuations are observed near the Dirac point. They are interpreted as the natural consequence of the presence of electron and hole puddles. The magnetic field at which the amplitude of the fluctuations is the largest is directly linked to the mean size of the puddles.

A few years ago, a new form of the integer quantum Hall effect (IQHE) was discovered in graphene for which the Hall conductance is half-integer quantized in units of \( 4e^2/h \) [1]. The factor 4 stands for Landau level (LL) degeneracy (twofold spin and twofold nodal degeneracy) so that the IQHE occurs at the filling factor \( \nu = 4 \times (|i| + 1/2) \), where \( i = 0, \pm 1, \pm 2 \ldots \) is an integer used for LL indexation. In clean graphene, close to the Dirac point (also referred to as the charge neutrality point (CNP)), spin and nodal degeneracies are expected to be lifted by a strong magnetic field. Indeed, pioneering experimental work [2, 3] has shown additional Hall resistance plateaus for the filling factor \( \nu = 0, \nu = \pm 1, \nu = \pm 4 \) as well as \( \pm 3 \) for which the role of electron–electron interactions has been particularly discussed [4]. The nodal degeneracy

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Figure 1. Longitudinal resistance as a function of gate voltage $V_g$ at $T = 1.5$ K and $B = 0$ T. The charge neutrality point is located at $V_g = +52$ V. The arrows indicate specific values of gate voltage at which magneto-resistance data will be presented (see figure 3). Inset: longitudinal resistance $R_{xx}$ and Hall resistance $R_{xy}$ as a function of magnetic field at $T = 1.5$ K and $V_g = 0$ V.

lifting implies spontaneous symmetry breaking effects [5, 6] whose fundamental nature is still under investigation [7]–[12]. It is now established that disorder significantly affects the electronic properties of graphene and thus cannot be ignored, especially at low charge carrier density, where it brings about the formation of electron–hole puddles [13]. Even if the field-induced insulator transition is well documented for clean graphene [14, 15], little is known about the high-field transport properties in disordered samples where the presence of electron–hole puddles is expected to play a significant role over a large magnetic field range [16].

In this paper, we report on IQHE in a disordered system for various charge carrier densities from hole to electron doping. At low field, our experimental data are consistent with existing theories for disordered graphene. Close to the Dirac point, the magneto-resistance is quasi-flat for a broad magnetic field range without sign of divergence in contrast with the clean case. In addition, at large magnetic field, we show that both longitudinal and Hall magneto-resistances display large fluctuations at the Dirac point, with maximum amplitude for $B \approx 25$ T. We show that these fluctuations result from the presence of electron–hole puddles with an estimated mean size of 5 nm.

The sample has been designed in the standard Hall bar geometry using electron-beam lithography, allowing simultaneous measurements of both Hall ($R_{xy}$) and longitudinal ($R_{xx}$) resistances. Variable electrostatic doping is achieved upon applying a voltage through 300 nm of SiO$_2$ dielectric used as a back-gate. Figure 1 shows the device resistance as the gate voltage $V_g$ is ramped from $-70$ to $+70$ V. The CNP is identified at the resistance maximum $V_{CNP} = +52$ V. The large full-width at half maximum (FWHM) of the curve as well as the large shift of $V_{CNP}$ from $V_g = 0$ V suggests a rather disordered sample. Non-intentional hole doping arises from defects and charged chemical species adsorbed at the surface of the sample or at the SiO$_2$ interface. The gating efficiency of the device is estimated to be $\beta = 7.85 \times 10^{10}$ cm$^{-2}$ V$^{-1}$.
from low-field Hall measurements. This result is compared to the accepted plane capacitor model for graphene $\beta = (\epsilon_r \epsilon_r)/(e d)$, where $\epsilon_r$ is the dielectric permittivity of SiO$_2$ and $d = 275$ nm its thickness, in good agreement with the nominal value. A constant current of $i = 0.5\mu A$ is injected through the device at 1.5 K, while a strong pulsed magnetic field, of total duration 300 ms and rising up to 57 T, is applied perpendicular to the sample. The inset of figure 1 shows IQHE at $V_g = 0$ V. The Hall resistance displays well-defined quantized plateaus at $R_{xy} = \frac{h}{4e^2} (i + 1/2)^{-1}$ for $i = 0, 1, 2 \ldots$ typical for graphene. The mean mobility is estimated to be $\mu = 1300 \text{ cm}^2/\text{V s}$, consistent with the first appearance of Shubnikov–de Haas (SdH) oscillations at minimum magnetic field $B^\dagger \approx 7$ T (see inset of figure 1).

For $B < B^\dagger$, in the classical regime, the experimental data can be approached using a standard two-fluid model [17, 18]. In this picture, electron and hole-like particles coexist and both contribute to transport. For simplicity and within a good approximation, we assume the same mobility $\mu$ for both electrons and holes. Their density will be denoted by $n_e$ and $n_h$, respectively. In the following, we use the reduced parameter $\alpha = (n_h - n_e)/(n_e + n_h)$ so that $R_{xx}(B) = R_{xx}(0) \times \frac{1 + (\mu \cdot B)^2}{1 + (\alpha \cdot \mu \cdot B)^2}$ and $R_{xy} = \frac{1}{\gamma} \mu \cdot B \cdot R_{xx}(B)$ with $R_{xx}(0) = \gamma \times [(n_e + n_p) \cdot e \cdot \mu]^{-1}$ and $\gamma = 3$ is the form factor [19]. A theoretical adjustment with $\alpha$ and $\mu$ as free parameters is performed to match the full experimental dataset. The agreement is convincing, as shown in figures 2(a) and (b). We emphasize the fact that $R_{xx}(0)$ is not a free parameter and is determined from the $R_{xx}(V_g)$ curve at zero magnetic field (see figure 1). Although some refinements (e.g. different mobilities for electrons and holes particles) would certainly improve the fitting quality, both the longitudinal and Hall resistances can be adjusted with this minimal two-fluid model that captures well the underlying physics of disordered graphene. This fitting procedure allows us to extract the profiles of $\alpha(V_g)$ and $\mu(V_g)$ (see figures 2(c) and (d)). On crossing the CNP, the hole-to-electron density ratio changes smoothly within a gate voltage interval $\Delta V_g \approx 20$ V. In the presence of disorder, mainly modeled by charged impurities, the coexistence of hole and electron-like particles is expected within a window $\Delta V_{g}^{th} = \frac{2n^*}{\beta}$, where $n^*$ is an effective charge carrier density defined by $n^* = \frac{(5 \times 10^{15}/\mu)^2}{4\beta V_{CNP}}$ [20]. We find $\Delta V_{g}^{th} \approx 23$ V, in good agreement with experimental results. The predicted minimum of conductivity at the CNP and zero field is $G_{CNP} = \frac{1}{\gamma} \times \frac{20e^2}{h} \times \frac{n^* \mu}{5 \times 10^{15}} = 61 \mu S$, which favorably compares with experiment (i.e. $G_{CNP}^{exp} = 60.6 \mu S$). In the first approximation, graphene’s resistivity was experimentally found to be inversely proportional to the density of charge carriers, so that their mobility is almost independent of $n \propto V_g$. This particularly holds for low-mobility samples, far from the Dirac point, where conduction is mainly dominated by long-range scatterers, such as charged impurities. On the other hand, little is known about the carrier’s mobility near the CNP since, when calculated using the simple Drude model, $\mu = \sigma / e \cdot n$ diverges for $n \to 0$. Using the two-fluid model fitting procedure, the extracted curve $\mu(V_g)$ in figure 2(d) shows a clear minimum of the mobility in the vicinity of the CNP. Do and Dollfus [21] compared the effects of long-range and short-range scattering centers on mobility. They demonstrated that, even if the mobility is peaked at $V_g = V_{CNP}$ in the presence of charged impurities, it is strongly reduced when vacancies are included, leading to a dip-like curve consistent with our observations.

We now turn our attention to high-magnetic-field transport properties. Figures 3(a) and (b) show the longitudinal and Hall resistances in the vicinity of the charge neutrality point. Focusing close to $V_{CNP} \approx 52$ V, the longitudinal magneto-resistance is first positive and reaches a maximum for $B \approx B^\dagger = 7$ T. This resistance maximum is attributed to the formation of overlapping LLs and naturally sets the crossover from the classical to the quantum regime.

Figure 2. (a, b) Theoretical adjustment of $R_{xy}(B)$ and $R_{xx}(V_g)$ at small field, respectively, at selected back-gate voltages or magnetic field using the two-fluid model. $R_{xx}(V_g)$ data have been successively upshifted to $\Delta R = 5 \, \Omega$ for clarity. Note that at $B = B^\dagger = 7$ T, the two-fluid model is no longer accurate. (c, d) Extracted coefficient $\alpha$ and mobility $\mu$ as a function of $V_g$: note the coexistence of electrons and hole-like particles for a broad range of gate voltages $\Delta V_g \approx 23$ V, as well as a mobility dip near CNP. Dotted lines are a guide for the eye.

The resistance then decreases as a consequence of the LLs energy interval increasing with magnetic field till the only $i = 0$ LL remains populated. From $B \approx 20$ T, a merely flat magneto-resistance is observed up to the highest experimentally available magnetic field. In graphene, the energy of the $n = 0$ LL is independent of magnetic field. Therefore, as long as the Fermi energy remains constant within the $n = 0$ LL, a fixed back-scattering rate between edge states through extended states accounts for the quasi-flat magneto-resistance. The scattering rate is maximum when $V_g = V_{\text{CNP}}$, that is, when the Fermi energy is aligned with the center of the $n = 0$ LL (see the inset of figure 3). As a matter of fact, instead of a purely flat magneto-resistance, a weak increase is observed at very high magnetic field. The closer $V_g$ is to $V_{\text{CNP}}$, the less pronounced the high-field magneto-resistance increase. This effect is explained considering a Fermi energy shift towards the center of the $n = 0$ LL as its degeneracy increases with the field. The energy shift of $E_F$ is all the more important as the initial density of states is weak. We note that, contrary to clean graphene devices [22], no divergence of the magneto-resistance is observed up to $B = 57$ T for this particular disordered device. The impact of disorder on our sample is expected to overcome the sample’s self-heating effects, which play only a minor role in this issue. This figure, together
Figure 3. Longitudinal resistance (a) and Hall resistance (b) as a function of magnetic field for selected gate voltages (see arrows in figure 1) in the vicinity of the charge neutrality point. Note that in (a) the curves have been successively down-shifted to $-5 \, \text{k\Omega}$ with respect to the curve at $V_g = 30 \, \text{V}$ for clarity. The inset shows $R_{xx}(B = 40 \, \text{T})$.

with the above-mentioned $R_{xx}(B, V_g)$ behavior, rules out any alternative explanations implying spontaneous symmetry breaking and degeneracy lifting that would occur in the whole sample. However, whether a divergent behavior will be detected or not for even higher magnetic field remains an open question. In the close vicinity of the CNP, the Hall resistance displays no more well-defined quantized plateaus (e.g. compared with $R_{xy}(B)$ curves at $V_g = 30 \, \text{V}$ showing a well-developed Hall plateau at $R_{xy} = 12.9 \, \text{k\Omega}$), but remains close to zero as a result of electron and hole balance. We note that, for some fixed values of the gate voltage, the Hall resistance experiences a sign inversion indicating a change in the majority carrier type. This effect is related to a Fermi energy shift of the system at high field and will be discussed in more detail elsewhere.

In the vicinity of the Dirac point, the Hall and longitudinal magneto-resistances are distorted with large fluctuations. Such fluctuating features are qualitatively analyzed in the conductance data, obtained by resistance tensor inversion. Figures 4(a) and (b) highlight such conductance fluctuations, where a smooth background function $\bar{G}(B)$ has been subtracted from the original conductance data. The standard deviation as a function of gate voltage is displayed in figure 4(c). The amplitude of the fluctuations is much larger in the vicinity of the CNP and in the intermediate magnetic field range ($10 \, \text{T} < B < 35 \, \text{T}$). Close to the CNP, large fluctuations have already been reported theoretically [23] and in various dc high-magnetic-field experimental works [2, 8, 11] as a function of gate voltage, but they are not systematically observed [24] depending on sample quality, magnetic field range and temperature. In disordered...
Figure 4. (a, b) Longitudinal and Hall resistance fluctuations for various gate voltages. A smooth background function has been subtracted from the original data. Data have been shifted for clarity. (c) Standard deviation of the curves shown in (a) (black) and (b) (red). (d) Maximum peak-to-peak amplitude of the fluctuations.

Materials, magneto-resistance fluctuations are usually understood in terms of magnetic-field-dependent phase shifts of the electronic wavefunctions and are well reproduced using the universal conductance fluctuations (UCF) theory [25, 26]. However, their occurring only in the vicinity of the Dirac point [27] and their large amplitude up to $2.5 \times 10^{12}$ e$^2$/h call for an alternative explanation. Following the lines of [20, 28], let disorder in graphene be described by a charged impurity density $n_{\text{imp}}$ that scales as $n_{\text{imp}} = 5 \times 10^{15} \mu^{-1}$. Using mobility $\mu = 1300 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for our sample, we compute $n_{\text{imp}} = 3.85 \times 10^{12} \text{ cm}^{-2}$, so that the typical mean scattering length is roughly $\ell \approx 5 \text{ nm}$. In disordered graphene, close to the Dirac point, the mean carrier density is low and screening is reduced so that the surrounding disordered medium drives the system into a fluctuating potential landscape, leading to the natural formation of electron and hole puddles. Using a scanning single-electron transistor, Martin et al. [13] have demonstrated that the mean puddle dimensions are of the same order as the typical disorder length. It is therefore reasonable to consider, at the CNP, an inhomogeneous system made of charged areas spread over the whole sample’s surface, defining electron-doped and hole-doped puddles that surround the sample’s charged impurities according to their specific local configuration. They may be irregular in shape but their minimum dimensions have a distribution centered on the mean distance between impurities ($\ell \approx 5 \text{ nm}$). When the magnetic field is increased, the magnetic...
length becomes smaller than the minimum puddle’s dimension, so that a cyclotron orbit can be accommodated inside the puddles. Consequently, the puddles successively enter the QH regime leading to magneto-resistance fluctuations with the maximum amplitude expected at $B \approx 25$ T, where $\ell_B = \sqrt{\frac{\hbar}{eB}} \approx \ell$. On the other hand, as the system is driven away from the CNP, the puddles become larger and the magnetic field required to fulfill the condition $\ell_B = \ell$ becomes smaller. This translates, in figures 4(a) and (b), to a maximum fluctuation amplitude shifted towards lower magnetic field as well as a weaker magnetic field range where fluctuations are visible. In addition, since the total number of puddles is decreased, the amplitude of the fluctuations is progressively reduced, as experimentally observed (see figures 4(c) and (d)).

To conclude, we have investigated the electronic properties of disordered graphene in the close vicinity of the Dirac point. At low magnetic field, the system is well described using a simple two-fluid model where both electron and hole-like particles contribute to transport. The coexistence energy interval is well accounted for by existing theory for disordered graphene, whereas the mobility dip at low carrier density is possibly explained by the presence of vacancies. When the magnetic field is high enough so that the energy spectrum displays a clear LL quantization, large fluctuations of the magneto-resistance near the Dirac point are observed and explained in terms of topological transition of electron–hole puddles into the quantum regime. These fluctuations progressively vanish as the magnetic field is further increased, suggesting a conduction regime where the magnetic field length is smaller than the mean puddle size. For clean graphene, spin and valley degeneracy splittings occur in strong magnetic field and, for the cleanest samples, the fractional quantum Hall effect [29, 30] has even been observed. Many experimental studies lead to the assumption that the magnitude of the magnetic field at which such effects occur in graphene is directly correlated to the disorder strength. There is no doubt that similar experiments with samples having different degrees of disorder will significantly broaden our understanding of the dirty regime.

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