Plateau–insulator transition in graphene

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**Abstract.** We investigate the quantum Hall effect (QHE) in a graphene sample with Hall-bar geometry close to the Dirac point at high magnetic fields up to 28 T. We have discovered a plateau–insulator quantum phase transition passing from the last plateau for the integer QHE in graphene to an insulator regime $\nu = -2 \rightarrow \nu = 0$. The analysis of the temperature dependence of the longitudinal resistance gives a value for the critical exponent associated with the transition equal to $\kappa = 0.58 \pm 0.03$.

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1. Introduction

The recent discovery of graphene [1], a flat monolayer of carbon atoms tightly packed into a two-dimensional (2D) honeycomb lattice, has attracted great attention, in particular after the experimental observation of a non-standard sequence of integer quantum Hall (QH) features [2, 3]. Graphene is the first truly 2D system available for investigation by solid state physicists. The unusual sequence of integer QH features is related to the quasi-relativistic nature of the charge carriers due to the particular graphene band structure near the Dirac point [4]. In graphene, the conventional integer QH quantization for the conductivity $\sigma_{xy}$ is shifted by a half-integer [4, 5] due to the half-filling of the $n = 0$ Landau level compared to the other levels:

$$\sigma_{xy} = \frac{4e^2}{h} \left( \pm n + \frac{1}{2} \right) = \frac{ve^2}{h}. \quad (1)$$

At very high magnetic fields and with high-quality samples, more QH plateaus not in the sequence of equation (1) have been observed [6]–[10]; these include filling factors $v = 0$, $\pm 1$, $\pm 4$ and the fractional quantum Hall effect has also been observed in suspended graphene samples [11, 12] with an insulating phase in the state $v = 0$ [11]. These new plateaus cannot be understood only by using the Landau quantization and their origin is currently a topic of considerable debate. Many theories attribute the origin of these new plateaus to some spontaneously broken symmetry of the system driven by electron–electron interactions (see [13] and references therein). Edge states could also play a key role and their relevance for the dynamics in graphene [7], [14]–[18] has been invoked to explain a variety of magnetotransport experiments near $v = 0$. Different measurements have demonstrated that the longitudinal resistance may either decrease [7] or increase [9] at $v \sim 0$ with decreasing temperature in samples that appear to be quite similar, fueling the debate concerning the existence and origin of an insulator phase near the Dirac point at high magnetic fields. This insulator phase has been attributed to field-induced spin-density waves in non-suspended graphene samples [19]. Our data show clearly a transition to an insulator phase at high magnetic fields being consistent with the results of Checkelsky et al [9] and Giesbers et al [10] but in a rather different regime, away from the Dirac point.

In the framework of the scaling theory of the integer quantum Hall effect (IQHE), plateau–plateau (PP) and plateau–insulator (PI) transitions are interpreted as quantum phase transitions with an associated universal critical exponent $\kappa$. The underlying physics is the Anderson localization–delocalization quantum phase transition [20]. In this work, we report experimental evidence for a PI transition between states at filling factors $v = -2$ and $v = 0$. We have obtained a value for the scaling exponent equal to $\kappa = 0.58 \pm 0.03$, to our knowledge, the first evidence of this exponent in a truly 2D system. The value we found for $\kappa$ is very similar to the values measured for this exponent for the PI transition in standard quasi-2D electron gases (2DEGs) in InGaAs/InP and InGaAs/GaAs quantum wells (see [21]–[26], [27] for a complete review).

2. Sample and magnetotransport measurements

We have used monolayer graphene flakes obtained by peeling graphite onto an Si wafer with a 300 nm SiO$_2$ top layer. They were processed in the CT-ISOM (Central de Tecnología-Instituto de Sistemas Optoelectrónicos y Microtecnología) clean-room facilities, depositing 50/500 Å
Figure 1. The left image shows the sample with the Hall bar covered by PMMA resist before the deposition of the contacts, while the right one shows the contacted sample with the Ti/Au already deposited.

Ti/Au contacts in a Hall-bar geometry by using e-beam nanolithography as can be seen in figure 1.

We made 12 Hall bars in different monolayer graphene flakes and we characterize them by magnetotransport measurements in our laboratory in Salamanca. We selected the sample S2601f4 that exhibited a high degree of homogeneity and the highest mobility. The Hall mobility (μ) for this sample ranges from μ = 4.5 × 10^3 cm^2 V^{-1} s^{-1} at a gate voltage V_{gate} = 17 V far away from the Dirac point to μ = 1.3 × 10^4 cm^2 V^{-1} s^{-1} at V_{gate} = 5 V and being 4.2 × 10^4 cm^2 V^{-1} s^{-1} near the Dirac point at V_{gate} = 1 V.

We have performed magnetotransport experiments in this sample up to 28 T at the Grenoble High Magnetic Field Laboratory using a 20 MW resistive magnet. On our first approach, we measured the density in a dilution fridge and the position of the Dirac point that was found at 3.8 V at T = 200 mK. The carrier concentration (electron and hole density in 10^{12} cm^{-2}) as a function of the gate voltage V_{gate} at B = 0 and 4.2 K is shown in figure 2. The gate voltage can be easily transformed into the carrier concentration given by

\[ n = \frac{C}{A} \left( \frac{V_{gate} - V_{Dirac}}{e} \right) \]

with C/A = 2.3 × 10^{-4} Fm^{-2} in the hole regime and C/A = 2.0 × 10^{-4} Fm^{-2} in the electron one (note the slight asymmetry in the obtained fitting curve), where C is the capacitance of the device (C ∼ 0.02 pF) and A its area (A = 107 μm^2).

The sample was then annealed for 90 min in a low-pressure He exchange gas atmosphere at 100 °C in situ prior to insertion into the 4He cryostat. We have observed a displacement of the Dirac point to −3 V (see figure 3(b)) becoming slightly narrower. Four probe measurements were carried out using standard ac lock-in techniques with excitation currents of 1–10 nA and frequencies of 1.3–13 Hz.

3. Results and discussion

In figure 3(a), the variation of the Hall (R_{xy}) and longitudinal (R_{xx}) resistances as a function of the magnetic field are shown. The temperature was 4.2 K and the gate voltage V_{gate} = −20 V. In such a configuration, we had the Fermi energy far away from the Dirac point with several Landau levels below it. At this gate voltage and keeping the temperature constant at 4.2 K, we extracted the hole density from the Hall measurements, being \( n_{2D} = 1.4 \times 10^{12} \text{ cm}^{-2} \) with...
Figure 2. Density in the electron and hole regime measured at 4.2 K (red dots) and interpolated function of the density (black line) for the sample prior to the annealing. As it can be seen, the Dirac point was at 3.8 V, while it was displaced to $-3$ V during the transport experiments.

Figure 3. (a) $R_{xx}$ (black line) and $R_{xy}$ (red line) as a function of the magnetic field in the hole-like region measured at 4.2 K for $V_{\text{gate}} = -20$ V. In this case, the density is $n_{2D} = 1.4 \times 10^{12}$ cm$^{-2}$ and the mobility $\mu = 4.5 \times 10^3$ cm$^2$ V$^{-1}$ s$^{-1}$. (b) $R_{xx}$ and $R_{xy}$ at $B = 0$ T as a function of gate voltage; the Dirac point is centered at $V_{\text{Dirac}} = -3$ V. The interpolated density of carriers measured at 4.2 K via the Hall slope (in $10^{11}$ cm$^{-2}$) is also shown for clarity.

$\mu = 4.5 \times 10^3$ cm$^2$ V$^{-1}$ s$^{-1}$. The Hall resistance displays plateaus with the expected sequence for single-layer graphene in accordance with equation (1). Figure 3(b) shows $R_{xx}$ and $R_{xy}$ as a function of the gate voltage without applying an external magnetic field. $R_{xy}$ at $B = 0$ shows a peak close to the Dirac point because $R_{xx}$ is much larger than $R_{xy}$ and even with a small Hall-contact misalignment the $R_{xx}$ contribution is dominant. The Dirac point is centered at $V_{\text{Dirac}} = -3$ V, remaining invariable in the following measurements shown in this paper. The interpolated density of carriers (obtained at 4.2 K using the same technique as in figure 2) is shown to extract easily its value compared with the position of the Dirac point.
Figure 4. (a) Longitudinal ($R_{xx}$) and Hall ($R_{xy}$) resistances at $T = 1.4, 2, 3$ and 4.2 K and (b) the conductivities $\sigma_{xx}$ and $\sigma_{xy}$ as a function of $B$ for $T = 2, 3$ and 4.2 K with $V_{\text{gate}} = -8$ V. The hole density is $n = 4.1 \times 10^{11}$ cm$^{-2}$ and the mobility is $\mu = 1.3 \times 10^4$ cm$^2$ V$^{-1}$ s$^{-1}$ at $T = 4.2$ K. In (a), the inset shows the Hall resistance for $+B$ and $-B$ in solid lines and $R_{xy}^u = \frac{1}{2}[R_{xy}(+B) - R_{xy}(-B)]$ in dashed line. In the inset of (b), we show the longitudinal $\sigma_{xx}$ and Hall $\sigma_{xy}$ conductivities as a function of the gate voltage at $B = 24$ T and at 4.2 K. $\sigma_{xx}$ shows a double-peak structure, indicating the splitting of the fourfold degeneracy. Meanwhile, the Hall conductivity shows a central plateau on which $\sigma_{xy} = 0$, with two lateral plateaus corresponding to $\sigma_{xy} = \pm 2e^2/h$.

Then we moved the gate voltage to $V_{\text{gate}} = -8$ V, close enough to the Dirac point to have only the lowest Landau level ($\nu = -2$) below the Fermi energy. Applying a magnetic field above 15 T we observed a transition to a new QH plateau ($\nu = 0$) not in the sequence of equation (1) as previously observed by other groups [6]–[9]. We focused our attention on the temperature dependence of this transition. In figure 4(a) we show $R_{xx}$ and $R_{xy}$ and in figure 4(b) the conductivities $\sigma_{xx}$ and $\sigma_{xy}$ (longitudinal and Hall) as a function of the magnetic field from 1.4 to 4.2 K, with $V_{\text{gate}} = -8$ V. The hole density was $n = 4.1 \times 10^{11}$ cm$^{-2}$ and the mobility $\mu = 1.3 \times 10^4$ cm$^2$ V$^{-1}$ s$^{-1}$. We observed a transition from the $\nu = -2$ to $\nu = 0$ state. The longitudinal resistance grew exponentially and appeared to exhibit a temperature-independent critical field at $B_c = 16.7$ T. The Hall resistance also has a temperature-independent critical point at $B_c = 16.7$ T and tended to the expected quantized value $\nu = 0$, reaching it at $B > 18$ T for at least 5 T. Unfortunately, the diverging behavior of $R_{xx}$ seriously hindered the determination of the Hall resistance $R_{xy}$, as even a small Hall-contact misalignment would result as the dominant contribution from $R_{xx}$. The traditional way to overcome this problem is by using the $B$ symmetry of the resistance components: $R_{xx}$ is expected to be symmetric when applying external $B$, while $R_{xy}$ is expected to be anti-symmetric [22]. The corrected $R_{xy}$ is obtained, by an anti-symmetrization of the measured $R_{xy}(+B)$ and $-R_{xy}(-B)$: $R_{xy}^a = \frac{1}{2}[R_{xy}(+B) - R_{xy}(-B)]$ measuring with reversed external magnetic field configurations ($+B$) and ($-B$) (both perpendicular to the 2DEG, ($+B$) being the standard configuration we have used in the work and ($-B$) the reversed magnetic field configuration; see the inset of figure 4 for clarity). In practice, because the admixture is a result of both a misalignment of contacts and a nonuniform current distribution in the sample, $R_{xy}$ itself is not entirely $B$-symmetric, limiting...
Figure 5. Longitudinal resistance ($R_{xx}$) as a function of $\Delta \nu$ with $\Delta \nu = 1/B - 1/B_c$ and $B_c = 16.7$ T at four different temperatures. In the lower inset, we show $\log(R_{xx})$ as a function of $B$. Using the standard scaling procedure we obtain $\nu_0$ by fitting our data to equation (2) and the results are plotted in the upper inset. We obtain a critical exponent $\kappa = 0.58 \pm 0.03$.

the accuracy to which $R_{xy}$ can be determined. Nevertheless, the corrected Hall resistance remained quantized deep into the insulating phase.

Our observations are likely to be understandable in terms of the usual picture for QH PI transitions. In a somewhat simplified picture of the standard QHE in 2DEGs, the transitions between plateaus take place at $\nu = \frac{3}{2}, \frac{5}{2}, \frac{7}{2}, \ldots$ etc. A special case is the transition from the last Hall plateau at $\nu = 1$ that occurs at $\nu = \frac{1}{2}$ where the 2DEG, instead of undergoing a transition between two plateaus, becomes an insulator. In the case of the PI transition in graphene, from $\nu = -2$ to $\nu = 0$ the expected filling factor at the transition is $\nu = -1$, almost the same as what we have obtained in our experiment $\nu(B_c) = -1.015$, confirming our interpretation of this transition.

In this study of the PI transition, we kept constant the carrier density (constant gate potential); then tuning $1/B$ was equivalent to tuning the filling factor. The experimental characterization of QH PP and PI quantum phase transitions relies on the behavior of the system as a function of $T$ for magnetic fields sufficiently close to a transition point. In the vicinity of this transition point the longitudinal resistance is expected to follow the empirical law [21, 26]:

$$R_{xx} = \exp \left[ -\Delta \nu / \nu_0(T) \right],$$

with $\Delta \nu = 1/B - 1/B_c$ ($\nu$ here should not be confused with the Landau level filling factor) and $B_c$ is the temperature-independent critical field set as 16.7 T as said before (left inset of figure 5). Fitting to equation (2) the experimental $R_{xx}$ measured at four different temperatures, we can obtain the associated critical exponent $\kappa: \nu_0 \propto T^\kappa$.

This approach has been used to study the PI transition in a great variety of quasi-2D systems. The seminal magnetotransport experiments of Tsui et al [20] in InGaAs/InP samples found a value for $\kappa$ of 0.42 in PP transitions. For a PI transition, although there remained some controversy, after two decades of experiments the most commonly measured value is $\kappa = 0.57$. 

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This value has been found for a vast range of samples and materials, also in agreement with numerical simulations [21]–[27].

In figure 5, we plot $R_{xx}$ as a function of $\Delta \nu$. We have not drawn possible maximum–minimum lines that can be extracted from upper- and lower-bound values because it will reflect the strength of the white noise, observed in magnetotransport experiments at very high magnetic fields (see for example [10]). We have fitted our data to the empirical law (2) with a typical least squares fit, and the goodness of single fit was testified by the fact that $R^2$ was about 0.97. We noted that the linear fit reproduces the behavior of the longitudinal resistance, and this indicates that the noise does not affect too much to the least squares accuracy. From these fits we obtained $\nu_0$ that are plotted in the right inset of figure 5 as a function of $T$. The exponent $\kappa$ was then obtained from $\nu_0 \propto T^\kappa$ the best fit being $\kappa = 0.58 \pm 0.03$ (with $R^2 = 0.99$). To obtain the overall incertitude we introduced the error of each single fit and temperature stability (10 mK).

We firmly believe that this is clear evidence of the existence of a QH insulator in graphene in the $\nu = 0$ state far enough from the Dirac point and that the transition from the last QH liquid ($\nu = \pm 2$) is the same (i.e. belongs to the same universality class) as the well-known PI transition in conventional quasi-2D systems.

Previous experiments on the zero-energy state in graphene in the presence of high magnetic fields were focused on the dependence on the gate voltage [7]–[9] (then for a fixed magnetic field) and close to the Dirac point. We observed that using the magnetic field as the driving parameter rather than the gate voltage, we can obtain a more reliable temperature-independent critical point. In previous studies, the $\nu = 0$ QH state did not show a clear plateau in $R_{xy}$, being only visible as a plateau in the Hall conductance [8, 9]. Our measurements, although fluctuating, show a plateau in the Hall resistance at $\nu = 0$. As we discussed above, the diverging behavior of the Hall resistance for magnetic fields above 24 T (see figure 4) is most likely due to Hall-contact misalignment. Unfortunately, our cryostat allowed us to vary the temperature only within a small range, that should be enlarged in future experiments. On the other hand, as it has been seen in previous experiments close to the Dirac point, the measurements showed large fluctuations and the nature of this behavior is not clearly understood [9]. Of course, we checked our results and in particular our fittings are stable and reproducible and our main results are not affected by these fluctuations.

Checkelsky et al [9] reported temperature-dependent transport measurements, where $R_{xx}$ exhibits a magnetic field-induced crossover to a state with a very large resistance, as in our measurements. However, they observed that for a fixed magnetic field the resistance was almost temperature independent below 2 K. This behavior, which appears to exclude a PI transition, was interpreted in terms of the symmetry breaking effect of the disorder potential, which may favor long-range order at finite temperature with a Kosterlitz–Thouless (KT) transition [9, 28]. In their case, the applied voltage was set extremely close to the Dirac point or even on it, while in our case it was 5 V away from it (note the big difference in terms of carrier density and mobility). They also observed how the divergence of $R_{xx}$ was shifted to higher fields when the Dirac point moved away from $V_g = 0$ V noting that it was important to choose samples with $|V_g| < 1$ V to investigate the intrinsic properties of the Dirac point, which is not our scenario.

We want to stress that our results are not in contradiction with the KT-like transition but rather probes a different regime. Away from the Dirac point, hence, when the magnetic field is varied in its strong regime (above 10 T the Zeeman splitting seems to be strong enough), the Fermi level is tuned through the first (spin-resolved) Landau level. Then the features reported in this work can be easily understood in terms of a transition dominated by percolation of the extended state.
The transition to an insulator reported in [9] occurs when the doping is set exactly at the Dirac point. Then, regardless of how strong $B$ is (as long as the spin is resolved), the Fermi level in the bulk is always precisely in the middle of the Zeeman gap. Due to particle–hole symmetry, the filling factor as well as $\sigma_{xy}$ and $R_{xy}$ vanish to 0. In this situation, the only gapless states that can in principle give rise to nontrivial dissipative transport are the ‘helical’ edge states discussed recently by Shimshoni et al [18]. In addition, we do not observe any trace of saturation over the range of temperatures investigated as in the previous papers [9]. Nevertheless, we would like to stress that a temperature-independent resistance has been observed in several experimental studies of PI quantum phase transitions. Shahar et al [21] reported for the first time this type of behavior in a number of different 2D systems and although it is not well understood it is usually related to some kind of long-range interactions [26] or to finite size effects [29]. In fact, finite size effects can be of particular relevance here due to the small size of graphene samples. Also, the observed fluctuations in the measurements of $R_{xx}$ and $R_{xy}$ pointed out in our data could be at least partially related to mesoscopic effects [30].

We therefore conclude that our data show clearly a PI transition from $\nu = -2$ to $\nu = 0$ most likely due to the presence of Anderson localized states. We stress, however, that this does not exclude that such a KT transition could exist for higher magnetic fields or when the doping was precisely set at the Dirac point being probable that both scenarios could coexist.

4. Summary and conclusions

Summarizing, we have studied the QHE in graphene and in particular the $\nu = -2$ to $\nu = 0$ transition. We observed a PI transition when the magnetic field is tuned across a critical field $B_c = 16.7$ T. Both the Hall conductivity and the Hall resistance remain quantized deep into the insulating phase, suggesting that we are in the quantized Hall insulator regime, the same that was observed for the PI transition in low-mobility 2D systems. Using the standard scaling theory analysis, we have obtained the critical exponent for this transition $\kappa = 0.58 \pm 0.03$, in agreement with the common value observed for this exponent in conventional 2D systems. This evidence supports the identification of $\nu = 0$ in graphene as an insulating phase rather than a QH state. To our knowledge this is the first measurement of this exponent in graphene and therefore the first measurement in a truly 2D system. Therefore two distinct PI transitions exist near the Dirac point in graphene, namely a conventional QH-insulator transition as the observed in our experiment and the recently observed KT transition by Checkelsky et al [9]. Further studies at lower temperatures and higher magnetic fields are needed to confirm our results in a larger range of temperatures and magnetic fields, and in particular to try to find the conditions where both transitions could be observed at the same time.

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