Quantum Hall effect in dual-gated graphene bilayers with tunable layer density imbalance

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We study the magnetotransport properties of dual-gated graphene bilayers, in which the total density and layer density imbalance are independently controlled. As the bilayer is imbalanced we observe the emergence of a quantum Hall state (QHS) at filling factor $\nu = 0$ evinced by a plateau in the Hall conductivity, consistent with the opening of a gap between the electron and hole bands. By varying the layer density imbalance at fixed total density, we observe a suppression of the QHS at filling factors $\nu = 8$ and $\nu = 12$ when the layer densities are balanced, an observation at variance with theoretical expectations in the absence of electron-electron interaction and disorder.

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Graphene, a layer of carbon atoms in a honeycomb lattice, has emerged in recent years as a new test-bed for electron physics in reduced dimensions [1, 2]. The linear energy band dispersion, zero energy band-gap, and chiral quasi-particles set this material apart from conventional two-dimensional electron systems (2DES) realized in semiconductor heterostructures. Graphite bilayers, consisting of two closely coupled graphene layers, are an equally interesting system with parabolic momentum-energy dispersion and chiral quasi-particles possessing $2\pi$ Berry phase [3]. Most interesting, in dual-gated graphene bilayers the electron on-site energy and density in each layer can be independently controlled, which in turn enables the band-gap energy tuning. Here we investigate the magnetotransport properties of dual-gated graphene bilayers with high-$k$ dielectrics, a device geometry that allows an independent control of the total density and layer density imbalance. As the bilayer is imbalanced we observe the emergence of a quantum Hall state (QHS) at filling factor $\nu = 0$, evinced by a plateau in the Hall conductivity, consistent with the opening of a gap between the electron and hole bands. By varying the layer density imbalance at fixed total density, we observe an unexpected suppression of the QHSs at filling factors $\nu = 8$ and $\nu = 12$ when the layer densities are closely balanced. This observation is at variance with the single particle picture, strongly suggesting that electron-electron interaction or disorder play a role in this system.

Our samples consist of natural graphite mechanically exfoliated [4] on a 300 nm SiO$_2$ dielectric layer, thermally grown on a highly doped $n$-type Si substrate, with an As doping concentration of $\sim 10^{20}$ cm$^{-3}$. Optical inspection combined with Raman scattering are used to identify graphene bilayer flakes for device fabrication. We define six metal contacts using electron beam (e-beam) lithography followed by 50 nm Ni deposition and lift-off [Fig. 1(a)]. A second e-beam lithography step followed by O$_2$ plasma etching are used to pattern a Hall bar on the graphene bilayer flake. A key issue for graphene gated devices is the deposition of a top dielectric without degrading the carrier mobility. Owing to the chemically inertness of graphite, attempts to grow high-$k$ dielectrics, such as Al$_2$O$_3$ or HfO$_2$, by atomic layer deposition (ALD) on clean highly oriented pyrolytic graphite leads to selective growth on terraces, where broken carbon bonds serve as nucleation centers. To deposit an Al$_2$O$_3$ top dielectric on our graphene bilayer samples, we first deposit a $\sim 20$ Å thin Al layer, which serves as a nucleation layer for the ALD of Al$_2$O$_3$. The sample is then taken out in air and transferred to an ALD chamber. Based on X-ray photoelectron spectroscopy and electrical measurements, the Al layer is fully oxidized thanks to the presence of residual O$_2$ in the Al evaporation chamber and the exposure to ambient O$_2$ [5]. Next a 15 nm-thick Al$_2$O$_3$ film is deposited using trimethyl aluminum as the Al source and H$_2$O as oxidizer. Lastly, a Ni top gate is deposited using e-beam lithography, metal deposition, and lift off [Fig. 1(a)]. The dielectric deposition technique used here has been shown to produce top-gated monolayer graphene samples with high, over 8,000 cm$^2$/Vs, carrier mobility [5]. The mobility of the dual-gated bilayer graphene samples investigated here is over $\approx 2200$ cm$^2$/Vs [7]. Longitudinal ($\rho_{xx}$) and Hall ($\rho_{xy}$) resistivity measurements are performed down to a temperature of $T = 0.3$ K, and using standard low-current, low-frequency lock-in techniques.

Hall measurements allow us to determine the total carrier density ($n_{tot}$) as a function of $V_{TG}$ and $V_{BG}$, as well as the corresponding capacitance values. For the sample investigated here the top- and back-gate capacitances are $C_{TG} = 225$ nF/cm$^2$ and $C_{BG} = 10$ nF/cm$^2$, respectively. A second parameter relevant for graphene bilayers is the layer density imbalance $\Delta n = (n_B - n_T)/2$, defined in terms of the difference between the bottom ($n_B$) and top ($n_T$) layer densities. The use of top- and back-gate allows us to independently control $n_{tot}$ and $\Delta n$. Up to an additive constant, $n_{tot}$ and $\Delta n$ are related to $V_{TG}$ and $V_{BG}$ by $n_{tot} = (C_{BG} \cdot V_{BG} + C_{TG} \cdot V_{TG})/e$, and $\Delta n = (C_{BG} \cdot V_{BG} - C_{TG} \cdot V_{TG})/2e$; $e$ is the electron charge. The definition of $\Delta n$ represents the layer density
imbalance in the limit of excellent screening in each layer, when the top and bottom gates control only the charge density in the top and bottom layers, respectively; the actual layer density imbalance is smaller than \( \Delta n \). The layer density imbalance translates into a transverse electric field on the graphene bilayer, \( E = e\Delta n/\varepsilon_0; \varepsilon_0 \) is the vacuum dielectric permittivity.

In Fig. 1(b) we show the sample longitudinal resistivity (\( \rho_{xx} \)) measured as a function of top (\( V_{TG} \)) and back (\( V_{BG} \)) gate biases, at a temperature \( T = 0.3 \) K. The diagonals of constant \( C_{BG} \cdot V_{BG} + C_{TG} \cdot V_{TG} \) represent the loci of constant \( n_{tot} \) and varying \( \Delta n \), while diagonals of constant \( C_{BG} \cdot V_{BG} - C_{TG} \cdot V_{TG} \) define the loci of constant \( \Delta n \) at varying \( n_{tot} \). The diagonal of \( n_{tot} = 0 \) is defined by the points of maximum \( \rho_{xx} \) measured as a function of \( V_{TG} \) at fixed \( V_{BG} \) values. In order to determine the \( V_{TG} \) and \( V_{BG} \) values at which \( n_{tot} = 0 \) and \( \Delta n = 0 \), we consider \( \rho_{xx} \) measured as a function of \( V_{TG} \) along the diagonal \( n_{tot} = 0 \) [Fig. 1(b) (top panel)]. As a function of \( V_{TG} \), \( \rho_{xx} \) displays a minimum and increases for both negative and positive \( V_{TG} \). This trend can be explained by the opening of a band gap in the graphene bilayer, as a result of different electron on-site energies on the two layers [8, 9, 10]. The band-gap increases with the transverse electric field and results in a reduced electrical conductivity [11, 12]. The \( \rho_{xx} \) dependence on \( V_{TG} \) along the diagonal \( n_{tot} = 0 \) allows us to determine the gate biases at which \( \Delta n = 0 \). The \( \rho_{xx} \) minimum on the \( n_{tot} = 0 \) diagonal of Fig. 1(b) (top panel) defines the \( \Delta n = 0 \) point [13]. Having established a one-to-one correspondence between \( V_{TG} \) and \( V_{BG} \) on one hand, and \( n_{tot} \) and \( \Delta n \) on the other, in the reminder of the manuscript we will characterize the bilayer in terms of \( n_{tot} \) and \( \Delta n \).

The dual-gated device of Fig. 1(a) allows independent control of \( n_{tot} \) and \( \Delta n \). In Fig. 2(a) we show \( \rho_{xx} \) vs \( n_{tot} \), measured at fixed values of \( \Delta n \), at a magnetic field \( B = 18 \) T and at \( T = 0.3 \) K. These data are measured by simultaneously sweeping \( V_{TG} \) and \( V_{BG} \), with the sweep rates adjusted such that \( \Delta n \) remains constant. The data shows QHSs, marked by vanishing \( \rho_{xx} \) at integer filling factors that are multiples of four [14]. This observation is explained by the four-fold degeneracy associated with both spin and valley degrees of freedom of each Landau level [3]. Using the measured \( \rho_{xx} \) and \( \rho_{xy} \), we determine the Hall conductivity (\( \sigma_{xy} \)) via a tensor inversion, \( \sigma_{xy} = \rho_{xy}/(\rho_{xx}^2 + \rho_{xy}^2) \). Figure 2(b) data shows the Hall conductivity (\( \sigma_{xy} \)) measured as a function of \( n_{tot} \), at \( B = 18 \) T and \( T = 0.3 \) K, and for different, fixed values of \( \Delta n \). Interestingly, the data of Fig. 2(a,b) reveals an increasing \( \rho_{xx} \) at \( n_{tot} = 0 \) with increasing \( \Delta n \), accompanied by the emergence of a plateau in the Hall conductivity at \( \sigma_{xy} = 0 \), which in turn indicates an emerging QHS at \( \nu = 0 \).

The emergence of a QHS at \( \nu = 0 \) in graphene bilayers can be explained by considering the electron energy levels in this system with and without an applied magnetic field. At \( E = 0 \) and \( B = 0 \) the graphene band structure has symmetric, parabolic electron and hole bands with a zero energy gap, and an effective mass \( m \approx 0.054m_0 \); \( m_0 \) is the free electron mass [3]. The electron and hole band are four-fold degenerate, owing to the spin and valley degrees of freedom. At finite \( E \)-field, or \( \Delta n \), the electron and hole bands are separated by an energy gap (\( \Delta \)) because of the different on-site electron energies on the two layers. In an applied perpendicular \( B \)-field the carrier energy spectrum consists of the four-fold degenerate Landau levels (LLs). At \( E = 0 \) an eight fold degenerate LL exists at energy \( \epsilon = 0 \), the electron-hole symmetry point. As a consequence of this degeneracy QHSs emerge at fillings that are multiples of four, and for \( \Delta n = 0 \), \( \sigma_{xy} \) exhibits a double \( 8e^2/h \) step across \( n_{tot} = 0 \) as shown in Fig. 2(b).

In an applied transverse \( E \)-field, the eight-fold degenerate LL at \( \epsilon = 0 \) splits into two, four-fold degenerate LLs.
These two LLs, containing electron and holes states respectively, are symmetrically positioned with respect to $\epsilon = 0$, and are separated by the same energy gap $\Delta$ which splits the electron and hole bands at $B = 0 \frac{\hbar}{e}$ In order to get further insight into the physics of the $\nu = 0$ QHS, in Fig. 2(c) we show $\rho_{xx}$ vs $\Delta n$, measured at $n_{tot} = 0$ and $B = 0$ T, along with the $\rho_{xx}$ vs $\Delta n$, measured at $B = 0$ and at $B = 18$ T. These data show that as a function of layer density imbalance, the $\rho_{xx}$ values at $\nu = 0$ in high $B$-field are comparable to the $\rho_{xx}$ measured at $n_{tot} = 0$ and $B = 0$, strongly suggesting that the $\nu = 0$ QHS and the energy band-gap opening at $B = 0$ as a results of layer density imbalance have the same origin.

Next, we examine the resistivity values measured as a function of the $B$-field, at fixed $n_{tot}$ and $\Delta n$. Figure 3 data shows $\rho_{xx}$ and $\rho_{xy}$ vs. $B$, measured at $T = 0.3$ K, at a total density $n_{tot} = 3.5 \times 10^{12}$ cm$^{-2}$, and for different $\Delta n$ values. Consistent with the four-fold degeneracy of the LLs, QHSs emerge at integer fillings that are multiples of four, namely $\nu = 4, 8, 12, 16$ etc... Examination of the Fig. 3 data reveals an interesting finding. The $\rho_{xx}$ measured at $\nu = 8$ and $\nu = 12$ is maximum at $\Delta n = 0$, and decreases with increasing $\Delta n$. This observation implies that the $\nu = 8$ and $\nu = 12$ are weakest at balance ($\Delta n = 0$), and become stronger as the bilayer is imbalanced. The dependence of the $\nu = 8$ and $\nu = 12$ QHSs as a function of charge imbalance is at variance with the single-particle theoretical picture\cite{8}, in which the LL spacing is independent of the applied transverse field, and consequently of $\Delta n$.

To further explore the unusual dependence on $\Delta n$ of the QHSs $\nu = 8$ and $\nu = 12$, in Fig. 4 we show $\rho_{xx}$ vs. $B$ measured at $n_{tot} = 3.5 \times 10^{12}$ cm$^{-2}$ for different values of $\Delta n$. Figure 4(a) shows the $\rho_{xx}$ vs. $B$ data measured for $\Delta n \geq 0$, when the carriers are transferred from the top to the bottom layer, while Fig. 4(b) shows the $\rho_{xx}$ vs. $B$ data measured for $\Delta n \leq 0$, when the carriers are transferred from the bottom to the top layer. The data of Fig. 4 substantiate our finding that $\nu = 8$ and $\nu = 12$ QHSs are weakest at balance and become stronger when the bilayer is imbalanced. A possible explanation for this finding would be a disorder asymmetry between the two layers, which may suppress QHS when the carriers reside predominantly in the more disordered layer. This is however ruled out by Fig. 4 data, which show that the $\nu = 8$ and $\nu = 12$ QHSs become stronger for both positive and negative $\Delta n$, roughly symmetric.
with respect to $\Delta n = 0$. Equally noteworthy is that the $\rho_{xx}$ vs. $B$ measured at $\Delta n = 0$ display a small oscillation pattern, reminiscent of universal conductance fluctuations. The $\rho_{xx}$ vs. $B$ data at $\Delta n = 0$ in panels (a) and (b) of Fig. 4 represent two different measurements, and show that the $\rho_{xx}$ oscillations are repeatable. These oscillations indicate edge-to-edge carrier scattering, consistent with the presence of extended electron states at $\nu = 8$ and $\nu = 12$ when these QHSs are weakest.

While the emergence of the $\nu = 0$ QHS as a function of charge imbalance can be explained by a single-particle picture as a signature of the opening of an energy gap as a function of an applied $E$-field, the dependence of the $\nu = 8$ and $\nu = 12$ QHS on $\Delta n$ cannot. Indeed, in this picture the LLs energy spacing is independent of $\Delta n$, which translates into QHSs that should be insensitive to an applied transverse $E$-field. We note however that this picture does not consider the role of disorder or electron-electron interaction. Disorder leads to LL broadening, which may also depend on the applied transverse $E$-field. A LL broadening which is minimum at $\Delta n = 0$ is consistent with the observed trend of Fig. 3 and 4. However, a theoretical study considering the role of disorder in graphene bilayers suggests that the QHSs strength in a disordered bilayer is independent of the applied $E$-field. Another mechanism that can explain the $\nu = 8$ and $\nu = 12$ QHSs dependence of $\Delta n$, is the electron-electron interaction in this system. While the role of electron-electron interaction on the LLs energy spacings in graphene bilayers has not been examined theoretically, Min et al. [10] showed that at $B = 0$ T the exchange interaction favors the spontaneous charge transfer between the layers, when a transverse $E$-field is applied.

In summary we report a magneto-transport study in a dual-gated graphene bilayer, in which the total density and charge imbalance are independently controlled. Concomitant with the energy band-gap opening at $B = 0$ in the presence of an applied transverse $E$-field, we observe the emerge of a QHS at filling factor $\nu = 0$, in agreement with single-particle theory [8]. Surprisingly, the $\nu = 8$ and $\nu = 12$ QHSs are suppressed when the bilayer is balanced, and become stronger when a transverse $E$-field is applied, rendering the bilayer imbalanced. This observation is at variance with the single-particle picture, in which the QHS energy gaps are independent of the $E$-field, and strongly suggests that electron-electron interaction or disorder play a role in stabilizing these QHSs.

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