Quantum Hall effect on centimeter scale chemical vapor deposited graphene films
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Citation: Appl. Phys. Lett. 99, 232110 (2011); doi: 10.1063/1.3663972
View online: http://dx.doi.org/10.1063/1.3663972
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We report observations of well developed half integer quantum Hall effect on monolayer graphene films of 7 mm × 7 mm in size. The graphene films are grown by chemical vapor deposition on copper, then transferred to SiO$_2$/Si substrates, with typical carrier mobilities $\approx$4000 cm$^2$/Vs. The large size graphene with excellent quality and electronic homogeneity demonstrated in this work is promising for graphene-based quantum Hall resistance standards and can also facilitate a wide range of experiments on quantum Hall physics of graphene and practical applications exploiting the exceptional properties of graphene. © 2011 American Institute of Physics. [doi:10.1063/1.3663972]
field effect mobility of \((3600 \pm 600) \text{ cm}^2/\text{Vs}\). A Hall mobility of \((4000 \pm 700) \text{ cm}^2/\text{Vs}\) at \(V_g = -30 \text{ V}\) is obtained by applying the same geometrical factor, consistent with the field effect mobility.

To further characterize the film quality and characteristic quantum Hall effect, we have measured \(R_{xx}\) and \(R_{xy}\) versus \(B\) (perpendicular magnetic field) at fixed \(V_g = -9 \text{ V}\) as shown in Fig. 2(b). The \(i = \pm 2\) quantum Hall plateaus are well resolved. The higher \(L_H\) plateaus are still not visible due to disorder induced \(L_H\) broadening. Below we focus on measurements by sweeping \(V_g\) at the highest available magnetic field, where better quality quantum Hall states are observed. The dc measurements are carried out in both four- and two-terminal configuration as a function of \(V_g\) at 1.7 K with \(B = 14 \text{ T}\) as shown in Fig. 2(a). The resistances are (anti)symmetrized between positive and negative magnetic field orientations to compensate for the imperfect geometry. The sharp \(i = 2\) to \(i = -2\) quantum Hall plateau to plateau transition happens within a gate voltage range of \((1.5 \pm 0.4) \text{ V}\) (measured between two extrema in the derivative \(d\sigma_{xx}/dV_g\) for \(n = 0 \text{ LL}\)), comparable with the transition for exfoliated graphene. \(^{18}\) This also confirms that the inhomogeneity in our large graphene film is small, as large inhomogeneity will tend to broaden the transition and even destroy the quantum Hall effect. The observed quantum Hall resistances are not exact, showing a residual \(R_{xx}\) of \(\approx 200 \text{ \Omega}\) for holes \((i = -2\) state\) and \(\approx 500 \text{ \Omega}\) \((i = +2\) state\) for electrons. \(R_{xy}\) also deviates from \(h/2e^2\) (the dashed horizontal line) by \(\approx 50 \text{ \Omega}\) for holes and \(\approx 100 \text{ \Omega}\) for electrons, qualitatively consistent with the theory that the absolute error in the quantization of \(\rho_{xy} (-R_{xy})\) can be related to a finite resistivity \(\rho_{xx}\) (indicating dissipation) as \(\Delta \rho_{xy} = -s \rho_{xx}\), where \(s\) is on the order of unity. \(^{19}\)

While the four-terminal resistance measurement is widely used to eliminate the effect of contacts and wires, two terminal triple-series connections proposed by Delahaye \(^{20,21}\) can also be used in both dc and ac bridges for high precision measurements of QHE. Assuming all of the contact and wire resistance values \(R_c, R_w \ll R_H = h/2e^2\), then from the dc equivalent circuit model, \(^{22}\) the resulting two-terminal resistance for a perfectly quantized \(i = \pm 2\) plateau is

\[ R = R_H \left(1 + 2 \left(\frac{R_c + R_w}{R_H}\right)^3\right). \]

Considering a few ohms of resistance presented in the wire, then \(R_c < 10 \text{ \Omega}\) is generally desired to achieve the precision of a few parts per billion when using \(R\) to measure the QHE. This is an experimental challenge for micron-sized graphene devices since the best normalized contact resistance so far is still \(\approx 500 \text{ \Omega} \mu\text{m}\). \(^{23}\)

Large contact resistance also introduces noise on the voltage probes and leads to local heating at the current contacts, thereby limiting the maximum breakdown current of QHE. The scalability of CVD graphene makes it possible to use large area contacts to reduce contact resistances. In our device, the contact size is \(\approx 1 \text{ mm}\); thus, \(R_c\) can easily fall below 10 \text{ \Omega} assuming a reasonable normalized contact resistance, i.e., \(\approx 5 \text{ k\Omega} \mu\text{m}\) for indium and clean graphene surface. While the precise determination of the contact resistance is difficult due to a non-vanishing \(R_{xx}\), the measured two terminal \(R\) agrees well with four terminal \(R_{xy}\) at the center of plateau, indicating that the contact resistance is likely much smaller than the residual \(R_{xx}\), and does not play a significant role here.

To investigate the reliability of this device, the sample is warmed up and exposed to atmosphere for 24 h and then cooled back down again. An ac lock-in technique is used for fast characterization. \(R_{xx}\) and \(R_{xy}\) are measured as a function of \(V_g\) at \(B = \pm 14 \text{ T}\) for different temperatures, and (anti)symmetrized as before (the gate sweep is performed over a wider range than that in Fig. 2, therefore revealing more quantum Hall states). We observe an increase in \(V_{\text{Dirac}}\) to 2.4 V and the QHE residual longitudinal resistance \(R_{xx}\) to \(\approx 600 \text{ \Omega} (\approx 750 \text{ \Omega})\)
for holes (electrons) at 1.7 K, indicates an increase of hole doping (and possibly disorder) from exposure to the atmosphere. A charge-neutral passivation layer similar to those reported for graphene on SiC (Ref. 24) may be employed in future to preserve the graphene quality in ambient environment. In any case, as shown in Fig. 3(a), the $i = \pm 2$ quantum Hall plateaus are still pronounced up to 60 K, showing the great potential as a quantum Hall resistance standard that can be used at much higher temperatures than those using GaAs heterostructures.

We have also characterized the zero magnetic field temperature dependent $R_{xx}$ for this device at different $\Delta V_g$ relative to the Dirac point as shown in Fig. 3(b), where $\Delta V_g = V_g - V_{\text{Dirac}}$, to get a qualitative indication of the disorder in the graphene. The resistance at the Dirac point increased by about 30% with decreasing temperature between 300 K and 1.7 K. For diffusive transport in the “dirty limit,” this temperature dependence is dominated by activation across potential barriers in inhomogeneous puddles.25 The rapid increase of resistance at temperatures less than 20 K is due to the weak localization. The temperature dependence becomes non-monotonic near $\Delta V_g \approx 4$ V, where the carrier density $n_i = 2.9 \times 10^{11} \text{cm}^{-2}$, similar to results reported in Ref. 26, showing that there still exist a fair amount of impurity scattering compared with exfoliated graphene or clean CVD graphene of very small size. At the high carrier density limit ($\Delta V_g = 50$ V) where the puddle effect is suppressed, $R_{xx}(T)$ exhibits metallic behavior, indicating scattering dominated by surface phonons. A better transfer technique is desired to best preserve the quality of as-grown CVD graphene.

In summary, we have studied the magneto-transport of large scale mono layer graphene grown by CVD on Cu, and then transferred to $\text{SiO}_2/\text{Si}$. In a 7 mm $\times$ 7 mm Van der Pauw geometry, these devices show half integer QHE at temperatures up to 110 K at $B = 14$ T, with a carrier mobility near 4000 cm$^2$/Vs. Such CVD graphene brings promising opportunities for graphene based integrated circuits, transparent electronics, quantum Hall resistance metrology, as well as optical and STM studies due to its exceptional electronic properties kept even at very large scale.

Y.P.C. acknowledges the support of NIST MSE grant 60NANB9D175. The authors would like to thank Shaﬁque Adam for discussions.

27. An empirical fitting to the field effect curve (Fig. 1) according to Kim et al. (Ref. 27) is used to calculate a geometry-independent residual carrier density $n_{\text{imp}} = 3.7 \times 10^{11} \text{cm}^{-2}$.
28. An empirical fitting to the field effect curve (Fig. 1) according to Kim et al. (Ref. 27) is used to calculate a geometry-independent residual carrier density $n_{\text{imp}} = 3.7 \times 10^{11} \text{cm}^{-2}$. From $n_{\text{imp}}$ and a theoretical model on graphene transport in the diffusive region by Adam et al. (Ref. 28) one can extract that the impurity concentration $n_{\text{imp}} = (1.4 \pm 0.2) \times 10^{12} \text{cm}^{-2}$ and a field effect mobility $\mu = (3600 \pm 600) \text{cm}^2/\text{Vs}$ assuming a typical distance of impurities from the graphene plane, $d = (1.0 \pm 0.2) \text{nm}$. This gives a geometrical factor (ratio between $R_{xx}$ and resistivity $\rho_{xx}$) of $4.2 \pm 0.7$ by comparing to the empirical fitting (Ref. 27) of the field effect curve.