Magneto-Transport Measurements on Multilayered Graphene on SiC(0001)

Mohammed Z Tahar
State University of New York, The College at Brockport, Brockport, NY 14420 USA
E-mail: mtahar@brockport.edu

Abstract. This report is on transport properties of a multilayer graphene on SiC(0001) sample. Both the magnetoresistance (MR) and Hall Effect measurements were carried out at ten constant temperatures (2 K - 300 K) and magnetic field scans 0 to 9 T. The results exhibit a negative MR and a sign reversal at low field (≤ 0.4 T) and temperatures above 50 K and no sign reversal below that up to $H \sim 2$ T. Additionally, for low temperatures (< 20 K) and high field (> 2 T), the MR signal shows SdH oscillations of the same frequency, $f = 65$ T. The Hall Effect signal shows a linear increase with magnetic field ($H < 0.3$ T) and a leveling off at higher fields. Using the two signals and the metallic resistance with residual resistance ratio ($RRR = 1.7$), one deduces the carrier concentration, $3.3 \times 10^{13}$ cm$^{-2}$, and mobility $3,700 - 21,000$ cm$^2$/V·s at 300 K and below 20 K, respectively.

1. Introduction

Graphene is one of the many allotropes of the element carbon and is a single sheet of 'graphite' that is one atom thick. Graphene is probably the ultimate 2D material, is important both from pure physics and device application point of view, and can be fabricated using three main processes: exfoliation (repeated pealing) of graphite [1], chemical vapor deposition (CVD) on metal substrates [2], and epitaxial growth on SiC [3, 4].

Epitaxial growth lends itself to micro-fabrication for possible electronics applications much better than both exfoliation and CVD. However, in epitaxial growth both the environment and the substrate have an effect on the final product as well as the number of layers and growth on SiC can be on either the Si or C terminated face, SiC(0001) and SiC(000T), respectively.

Graphene’s exotic properties are due to its 2D hexagonal structure, which leads to a linear energy-momentum relationship, that gives rise to massless Dirac Fermions and valley and chiral symmetries. Experimentally, these properties are known to be dependent on the growth methods and substrates used, as well as the number of layers and their stacking correlation. Magneto-transport studies can shed some light on the characterization of these intricate dependencies. Graphene physical properties are reviewed in the article by Castro Neto et al [5], where one can find an extensive theoretical treatment of single and multi-layer graphene of finite sheet size. For treatment of magneto-transport in graphene one is referred to the Physical Review Letter by McCann et al. [6].

This is a report on magneto-transport measurements on a sample of graphene of six layers and 2µm width grown on SiC(000T) in the temperature range 2 K - 300 K and in magnetic fields up to 9 T. The results are analyzed in light of the above references and compared to others in the literature.
2. Experimental Details

Graphene was grown by low pressure sublimation in an inductively heated quartz tube furnace. The SiC(000\text{1}) substrate was placed in a graphite box, and pumped overnight with a turbomolecular pump. For this particular sample, growth was performed in 50 mbar of Ar flowing at 5 standard liters per minute (SLPM). The temperature was ramped from room temperature to a growth temperature of 1525 °C, and held for 25 minutes. After growth was completed, the power supply controlling the heater was shut off, and the pressure was ramped to 900 mbar. The elevated Ar pressure impeded the growth of additional graphene as the reactor cooled. Full details of the growth process, the number of layers determination using XPS and optical measurements, and the absence of layer stacking correlation using low energy electron diffraction (LEED) have been described and discussed by Robinson et al. [4].

The transport measurements were carried out on the graphene sample using the multi-point contacts method. The contacts on the sample were made using lithography to larger pads onto which gold was wire-bonded, as shown in Fig. 1. The thin and short gold wires were connected to stronger copper wires, for soldering onto the sample holder pads, using a two-component silver loaded epoxy which was cured at ambient temperature for 24 hrs. Because these different metal-metal joints can give rise to thermoelectric voltages that depend on the metals, the temperature, and its gradient across the leads, an AC excitation from a floating current source [7] was used, along with lock-in-detectors at low frequency. At 7.000 Hz the frequency is low enough for the measurements to be considered DC measurements. To avoid the sample’s self heating, the excitation current (< 10μA) was so that the total power dissipation was under 0.01µW. The sample holder was inserted in a Quantum Design’s Physical Properties Measurement System (PPMS) cryostat with a superconducting magnet capable of ramping the magnetic field at 0.6 T/min. up to 9 T and sample’s temperature control for setting and holding with reproducibility < 1% or better, all in high vacuum. The applied magnetic field is perpendicular to the basal plane of the graphene sample and the probing current is parallel to it.

![Figure 1](image1.png)

**Figure 1.** Optical image of contacts to the sample and the bond-pad layout. The current is right to left, the magnetic field is out of the plane of the image.

![Figure 2](image2.png)

**Figure 2.** Magnetoresistance, $\frac{\Delta R(H)}{R(0)}$, with field (in Tesla) scans at different temperatures (2K - 300K) as indicated by the legend on the right of plot.

Referring to Fig. 1, the current was injected from the right middle pad and returned through the left pad, the longitudinal (magnetoresistance) voltage leads were connected to the top two outer pads, and the transverse (Hall Effect) voltage contacts were connected to the middle pads, with the top as positive and the bottom as negative. Notice the Hall contacts are not directly
facing each other. The applied magnetic field is perpendicular and coming out of the plane of the image. The measurements were carried out as follows. The PPMS computer set and maintained a stable sample temperature and ramped the magnetic field up and down to the set field values, while another PC collected the different voltages off of two lock-in-detectors, one for the Hall voltage and the other for the resistance voltage.

3. Results
Figure 2 shows all the magnetoresistance, \( \frac{\Delta \rho(H)}{\rho(0)} = \frac{\Delta R(H)}{R(0)} \), results for field scan-ups (0 to 9 T) at discrete temperatures of 300, 200, 100, 50, 20, 10, 5, 4, 3, and 2 K, as indicated by the legend on the right hand of the graph. One can distinguish two behaviors of the magnetoresistance with field at the different temperatures. First, a high temperature \( T > 50 \) K field dependence, where the magnetoresistance dips below zero for low fields, goes through a minimum at \( H_{\text{min}} \), rises above zero to reach a maximum, and then slowly tapers off below zero for higher fields. This behavior is clearly seen in Fig. 3, which is a graph of the same MR data of \( \frac{\Delta R(H)}{R(0)} \) vs. \( H \), but only up to 2 T for more details.

The first zero crossings (at \( H_1 \)) from negative to positive magnetoresistance, occur in the field range \( 0.2 < H < 0.3 \) T and the second and reverse crossings (at \( H_2 \)) occur in the range \( 1.3 < H < 2.1 \) T. The magnetic fields at which the first crossings (\( H_1 \)) occur seem to display a weak temperature dependence for \( T > 50 \) K and more than quadrupling for the temperatures below that. This weak temperature dependence is also mimicked by the field values at which the minima occur. This is shown in Fig. 4, as a plot of \( H_{\text{min}} \), \( H_1 \), and \( H_2 \) as functions of temperature. The magnetoresistance also displays a broad maximum at \( H \sim 0.8 \) T, with similar but much weaker temperature dependence.

Second, at low temperatures \( T < 20 \) K, there is a distinct field dependence, where the data dips below zero for low fields, goes through a minimum, and then rises but remains mainly below zero up to high fields, where Shubnikov-de Haas (SdH) oscillations are evident.

Figure 5 shows all the results for the Hall Effect voltage as a function of applied magnetic field at different temperatures. Due to the misalignment of the Hall contacts (see Fig. 1) a magnetoresistance correction was applied. In Fig. 5, the Hall Effect voltage is always positive.
Figure 5. The corrected Hall Effect voltage with field scans at different temperatures (2K - 300K).

Figure 6. SdH oscillations of MR signal vs. $\frac{1}{H}$ ($T^{-1}$) for $T < 20$ K. The inset is a plot of the Landau Level index, $n$ vs. $\frac{1}{H}$; its slope gives the frequency of oscillations.

for all temperatures, thus determining the carriers’ positive polarity. One can distinguish the linear dependence of the Hall voltage on the applied field below 0.4 T for all temperatures, and subsequent different saturation/levelling off behavior for the high temperatures ($T > 50$ K). The Hall voltage linear region is used to obtain a slope, which in turn is used to infer the carrier concentration at that temperature. Further, the carrier concentration, the sample’s measured resistance at that temperature and $H = 0$ T, the width, and the distance between the contacts are used to calculate the mobility of the carriers.

Figure 7. $R(T)/R(300K)$, with a room temperature resistance of 36Ω.

Figure 8. Carrier concentration, $n(T)$, in $10^{13} \text{ cm}^{-2}$ (blue) and mobility, $\mu(T)$, in $10^4 \text{ cm}^2/\text{V} \cdot \text{s}$ (red).

Figure 6 shows the low temperature ($T < 20$ K) behavior of the magnetoresistance and its Shubnikov-de Haas oscillations, as $\frac{\Delta R(H)}{R(0)}$ vs. $\frac{1}{H}$. As can be seen, there are definite oscillations in $\frac{1}{H}$. However, their weakness, which may be due the micro width of the sample, prevents
performing a Fourier transform on them to obtain the frequency(s). Instead one can label the maxima (peaks) as Landau levels and obtain the \( \frac{n}{\pi} \) value at which they occur, as in Wu et al. \[8\]. This results in the inset of Fig. 6, as a plot of the Landau level index, \( n \), as a function of \( \frac{1}{\pi} \). A linear least squares fit to that data set yields a frequency of 65 T and an intercept of 0.21 ± 0.34, which may be indicative of the anomalous Berry’s phase.

Figure 7 shows the resistivity temperature dependence in a semi-log plot of \( R(T)/R(300K) \) vs. \( T \). The sample has a room temperature resistance of 36Ω and a residual resistivity ratio (RRR) of 1.7, as can be deduced from the graph. This graph clearly shows the metal-like behavior of the resistivity of the sample. The resistivity of the sample is reasonably constant at low temperatures, up to 50 K, and increases with temperature above that, which is consistent with Matthiessen Rule.

Figure 8 shows a semi-log plot of the resulting carriers’ concentration, \( n \), in units of \( 10^{13} \text{ cm}^{-2} \) (blue) and their mobility, \( \mu \), in units of \( 10^4 \text{ cm}^2/\text{V-s} \) (red) as functions of temperature, on the same graph. Both the concentration and the mobility of the carriers exhibit a metal-like behavior. They are both constant for temperatures below 50 K, and the carriers concentration subsequently increases with temperature due to thermal excitation above 50 K while the mobility decreases due to phonon scattering, as the phonons population increases with temperature.

4. Discussion/Conclusion

According to Hass et al. \[9\], multilayer graphene on SiC(000T) behaves like a single sheet graphene. The behavior of the magnetoresistance results seem to qualitatively agree with theoretical prediction(s) by McCann et al. \[6\] as given in their Fig. 2, especially for field \( H < 2 \text{ T} \). The two predicted regimes correspond to the low temperature (\( T < 20 \text{ K} \)) behavior before the onset of the SdH oscillations and the high temperature (\( T > 50 \text{ K} \)) behavior. The characteristic magnetic fields (\( H_{\text{min}} (T) \) and \( H_{\text{f}} (T) \)) and their temperature dependence in Fig. 4 may be linked to characteristic scattering times associated with phase-coherent graphene \[6\] by \( H = \frac{h}{\pi \tau} \), with \( D \) as the carrier diffusion constant.

The period of the SdH oscillations, \( \frac{1}{f} = \Delta (1/H) \), is related to the area of an extremal orbit \[10\] by:

\[
A_e = \frac{4 \pi^2 e}{h} / \Delta (1/H) = \frac{4 \pi^2 e}{h} f
\]

where \( e \) is the electronic charge and \( h \) is Plank’s constant. Here, the orbit is a circle whose radius is \( k_F \), the Fermi momentum, and whose area is \( A_e = \pi k_F^2 \). From the theoretical review of Castro Neto et al. \[5\], the Fermi momentum and the carrier concentration are related by \( k_F^2 = \pi n \). \( n \) is the carrier concentration, even when the carrier polarity is positive.

All this in turn yields the carrier concentration as \( n = \frac{4 \pi f}{e} \). Using the SdH frequency 65 T, the result is \( n = 6.3 \times 10^{12} \text{ cm}^{-2} \), which compares favorably to \( 9.2 \times 10^{12} \text{ cm}^{-2} \), obtained from an average of the Hall Effect measurements at low temperatures. These carrier concentration values are comparable to the lower values obtained by Wu et al. \[8\] and consistent with the number of graphene layers of the sample. Additionally, the low temperature mobility values are lower but comparable to the results by Berger et al. \[11\] on epitaxial graphene, which may be due to the high carrier concentration deduced from the Hall Effect measurements.

Further, considering the residual scattering, one can infer the carrier mean free path and compare to the width of sample. Using the relation between the conductivity, mobility, and mean free path, \( \sigma = e n \mu = \frac{2e^2}{h} (k_F l) \), one obtains the mean free path, \( l = \frac{h}{2e} \sqrt{\frac{2}{\pi \mu}} \). Use of the average numerical values for \( n \) and \( \mu \) at low temperature yields 1\( \mu \text{m} \), a factor of two smaller than the width of the sample.
Acknowledgment
The author is grateful to Dr. Z. R. Robinson (SUNY Brockport Physics) and Dr. A. Nath (Navy Research Lab. Washington, DC) for providing the sample as well as fruitful discussions, and to the Department of Physics for local institutional support.

References
[1] Kovoselov K S, Geim A K, Morozov S V, Jiang D, Zhang Y, Dubonos S V, Grigorieva I V, and Firsov A A, Science 306, 666 (2004).
[2] Winterlin J and Bocquet M L, Surf. Sci. 603, 1841 (2009).
[3] de Heer W A, et al., Solid State Commun. 143, 92 (2007).
[4] Robinson Z R, et al., Carbon 81, 73 (2015).
[5] Castro Neto A H, Guinea F, Peres N M R, and Novoselov K S, Rev. Mod. Phys. 81, 109 (2009).
[6] McCann E, Kechedzhi K, Fal’ko V I, Suzuura H, Ando T, and Altshuler B L, Phys Rev. Lett. 97, 146805 (2006).
[7] Burin P P and Pfleiderer C, Rev. Sci. Inst. 67, 4024 (1996).
[8] Wu X, Li X, Song Z, Berger C, and de Heer W A, Phys Rev. Lett. 98, 136801 (2007)
[9] Hass J, et al., Phys Rev. Lett. 100, 125504 (2008).
[10] Kittel C, Intrduction to Solid State Physics, 2005 (Wiley & Sons).
[11] Berger C, et al., Science 312, 1191 (2006)