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Charge transport in CVD graphene

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Abstract

Graphene is a mono-layer of sp^2 bonded carbon atoms arranged in a honeycomb pattern. It is considered a promising candidate for the next generation of electronic materials as it exhibits both electron and hole conduction with extremely high carrier mobilities. Graphene films were prepared via chemical vapor deposition and then transferred to doped Silicon/Silicon dioxide (Si/SiO_2) wafers. It was washed in toluene several times to remove any organic deposits left behind during the transfer process. Atomic force microscopy images shows that the graphene surface is very rough. The graphene device was then electrically characterized in a field effect transistor configuration as a function of temperature and the resistance calculated at the Dirac point minimum of the device transconductance curve. The Dirac point resistance showed weak temperature dependence characteristic of graphene. Thermally evaporating 1nm of silver (Ag) over the device reduced the surface roughness and made the graphene surface more planar. Electrically characterizing the device again showed that there was reduced mobility but no change in the charge transport mechanism. Our results imply that the charge mobility in graphene is largely governed by the contact with the substrate and the charge transport mechanism is unaffected after covering the surface with 1nm of silver.

Keywords: Graphene, Mobility, Temperature

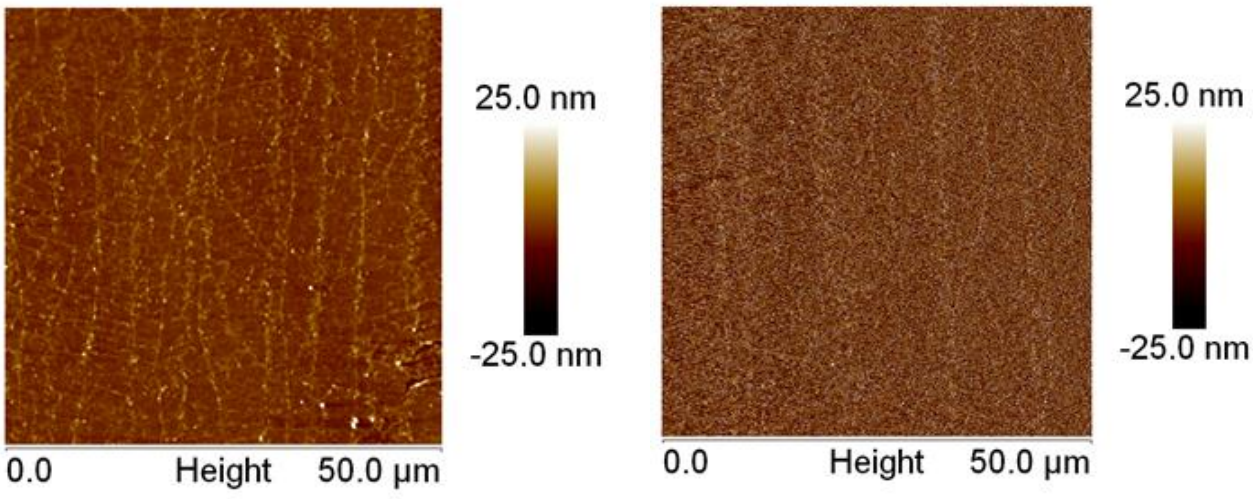
1. Introduction

Graphene is a mono-layer of sp^2 bonded carbon atoms arranged in a honeycomb pattern and can be considered as an ideal two dimensional (2-D) electronic system[1]. The charge carriers in graphene reside in the valence and conduction band which touch at two independent points, K and K' at the border of the first Brillouin zone. The large current measured in graphene is contrary to the results of scaling theory where all states are localized in 2-D systems and hence are insulators[2]. Quantum interference effects suppress the backscattering of charge carriers in the honeycomb crystal structure and are responsible for the observed high conductivity. Typical sheets of graphene show metallic like behavior of charge transport i.e. the temperature dependence of the resistance is independent or very weak in nature. Introducing external defects via hydrogen substitution or metal cluster deposition leads to scattering and a transition from the metallic to the insulating regime where variable range hopping is the dominant charge transport mechanism.

Although initially obtained via exfoliation of graphite, graphene can now be grown in macroscopic sizes (>1 cm^2) via chemical vapor deposition (CVD). The transfer of graphene made via CVD from the copper foil on which it is grown to the silicon substrate leads to a rough topography with many grain boundaries that affect the charge mobility. In order to smoothen out the rough graphene surface, we look at the effects of coating it with a thin silver film. By measuring the temperature dependence of the Dirac point resistance of the graphene film, we find that there is no change in the charge transport mechanism although the mobility of the silver (Ag) coated film decreases.

2. Experiment

Graphene was prepared via chemical vapor deposition (CVD) at the University of Pennsylvania. After transferring the graphene film onto a doped Si/SiO_2 substrate, it was washed several times with toluene to remove any organic material left behind during the transfer process. A Transmission Electron Microscope (TEM) grid was then placed over the graphene surface and was used as a template to fabricate electrodes for external electrical connections. A high vacuum evaporator was used to evaporate 100 nm of Ag over the grind and the substrate. After removing the TEM grid, a pattern of electrodes were found to be deposited on the graphene surface. Two contact pads were electrically contacted with gold wire and silver paint and formed the source and drain electrodes. Another contact to the backside of the wafer constituted the gate electrode. For electrical characterization, the device was placed in a closed chamber under a vacuum of 10^{-2} Torr. The source-drain voltage and currents were supplied and measured with a Keithley Model 6517B electrometer and the gate voltage was supplied with a Keithley Model 6487 pico-amp meter/voltage source.



(a) (b)

Figure 1. Atomic Force Microscope (AFM) image of the graphene surface (a) before and (b) after the deposition of 1nm Ag.

The as received graphene has a rough surface as seen in Figure 1(a), this is due to the transfer process to the Si substrate. It involves the use of harsh chemicals to dissolve copper and other polymer films used in the transfer process. In order to reduce this surface roughness, we evaporated 1nm of Ag over the graphene surface. Figure 1(b) shows an AFM image after this deposition where the surface appears to be smoother although faint lines of the underlying structure can be barely seen.

3. Results And Discussion

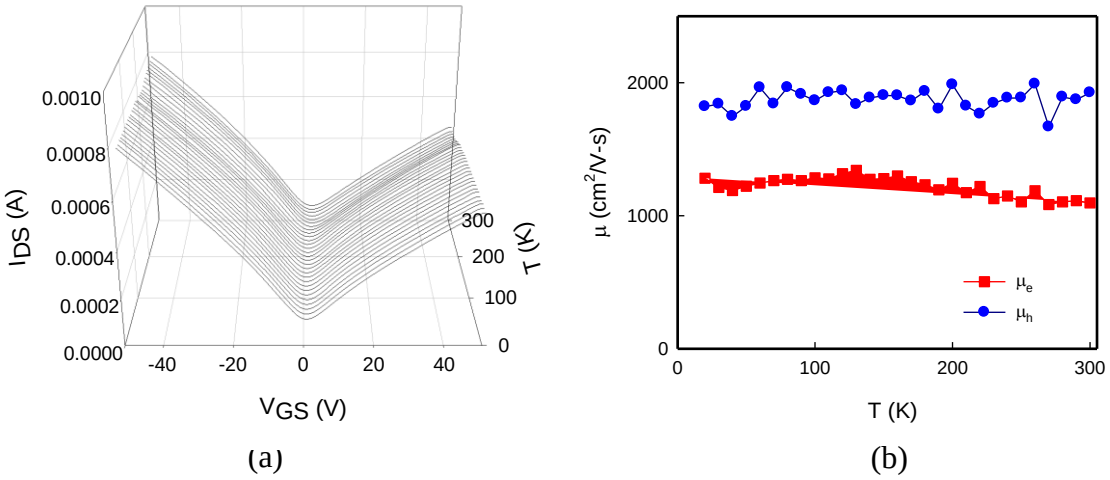


Figure 2: (a) Device current (I_{DS}) vs. back gate voltage (V_{GS}) as a function of temperature before Ag coating. (b) Temperature dependence of the hole (μ_h) and electron (μ_e) mobility extracted from the curves in Figure 2(a).

Figure 2(a) is a three dimensional plot of the device current expressed in units of amperes, versus gate voltage (V_{GS}) for an as-fabricated CVD graphene device measured under a vacuum of 10^{-2} Torr as a function of temperature (T). From this plot the device transconductance (gm) is calculated using equation (1):

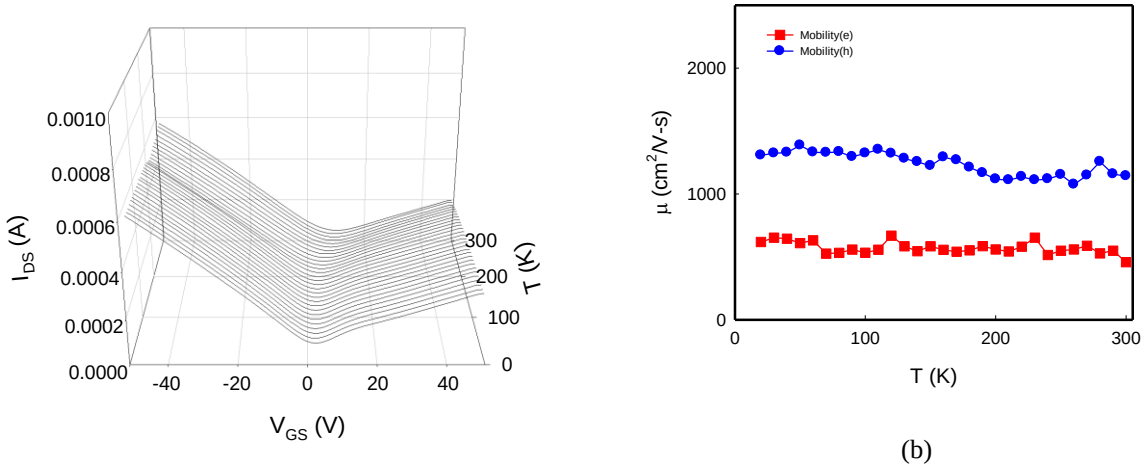
$$g_m = \left[\frac{\partial I_{DS}}{\partial V_{GS}}\right]_{V_{DS}=constant} \quad (1)$$

The I_{DS}-V_{GS} plot shows a typical "V" shape that is characteristic of graphene, with enhanced current in the hole-branch (negative V_{GS}) compared to that in the electron-branch (positive V_{GS}); similar observations on exfoliated graphene were attributed to different contact resistances for hole carriers[3]. As one moves away from the minimum point on this curve (the Dirac point) for each temperature, the current increases linearly with V_{GS} for both carrier polarities, indicating that the mobility is independent of carrier concentration and type, in agreement with measurements of exfoliated graphene, as well for graphene produced via CVD[4]. At gate voltages further from the Dirac point, the current deviates from linearity, most likely due to the presence of charged impurities or water underneath the graphene surface. The fact that at V_{GSmin}, the value of V_{GS} at the Dirac point exhibits a non-zero value, along with the fact that the width of the minimum current near the Dirac point is broad, indicates the existence of disorder and the presence of charge impurities. From this plot the charge mobility can be calculated from equation (2):

$$\mu = \frac{g_m L}{W C_i V_{DS}} \quad (2)$$

where g_m is the device transconductance, L is the channel length, W is the channel width and C_i is the capacitance per unit area of the silicon oxide surface below the graphene. V_{DS} is the drain-source voltage that was held fixed at

50mV. The slopes of the linear portion of the curves to the left and right of the Dirac point yield g_m for holes and electrons respectively, from which the mobility of each charge carrier species can be calculated. Figure 2(b) shows the dependence of μ as a function of temperature for electrons and for holes. The mobility is seen to be fairly constant in the temperature range 20K< T< 300K. The hole mobility is also seen to be higher than the electron mobility for all measured temperatures.



(b)

Figure 3. (a) Device current (I_{DS}) vs. back gate voltage (V_{GS}) as a function of temperature after Ag coating. (b) Temperature dependence of the hole (μ_h) and electron (μ_e) mobility extracted from the curves in Figure 3(a).

Figure 3(a) is a plot of the device current I, expressed in units of amperes, versus gate voltage (V_{GS}) after the device was coated with 1nm of Ag and Figure 3(b) shows dependence of the mobility as a function of temperature for electrons and for holes. The characteristic behavior of the curves in Figure 3(a) is similar to that in Figure 2(a), but now both the hole and the electron mobility have decreased compared to that shown in Figure 2(b). When Figures 2(a) and 3(a) are compared we note several differences: (i) At 300K, the gate voltage at the Dirac point is further to the right (toward positive gate bias) after the device is coated with Ag. (ii) Lowering the temperature shifts the gate voltage at the Dirac point towards the left (i.e. toward *n*-doping). This implies that the silver coating shows larger *n*-doping effect and is due to the electron donating feature of silver. The lower mobility indicates that the silver coat, even though it smoothens the graphene surface, has several scattering centers on the graphene film that impede the flow of charge in the graphene channel.

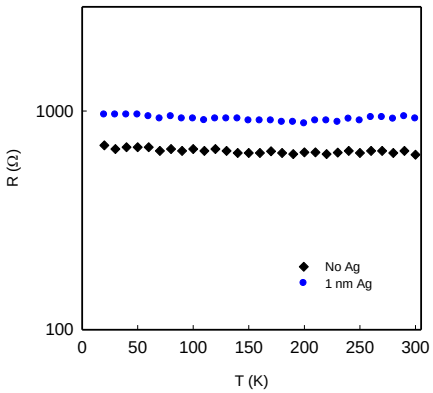


Figure 4. Temperature dependence of Dirac point Resistance before Ag coating (♦) and after Ag coating (•).

Figure 4 shows the temperature dependence of the resistance (R) calculated at the Dirac point, R= $\frac{V_{DS}}{I_{DSmin}}$, before and after coating with 1nm Ag. We can see that when the device has a 1nm Ag coating the resistance is much higher than without Ag coating. With Ag coating the surface of graphene gets smoother however it exhibits lower Mobility. On the other hand, without Ag coating the surface of graphene is rough but the resistance is lower. We can see that no matter the change of temperature the resistance is relatively constant i.e the slopes of the curves in Figure 4 are similar. This is an indication that the charge transport mechanism is not altered after Ag deposition. From this Figure we also see that the dependence with temperature is weak and consistent with the highly conducting nature of graphene.

4. Conclusions

CVD graphene was electrically characterized in a field effect transistor configuration as a function of temperature before and after coating it with a thin layer of Ag. The silver coating made the top surface of graphene smooth; this however reduced the hole and electron mobility implying that it also acted as scattering centers toward charge transport. The temperature dependence of the resistance did not change with the Ag coat. This implies that the charge transport mechanism is not sensitive to the top surface of graphene, instead it is governed by the bottom contact of the graphene with the SiO_2 surface.

5. Acknowledgments

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6. References

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