Metal-Insulating transition in disordered graphene nanoribbons controlled by helium ion irradiation

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Abstract

We report a mobility transition in graphene nanoribbons subject to disorder induced by irradiating helium ions. At small irradiation doses defects are created in graphene, enhancing scattering and decreasing the mobility. As the dose is increased further an abrupt transition into an insulating phase is observed characterised by a decrease in electron mobility by more than 5 orders of magnitude. Transmission Electron Microscopy images reveal that heavily He⁺ irradiated graphene loses its crystallinity as carbon atoms are dislodged from their position by incident ions and reconstruct into an insulating, topologically disordered two-dimensional sp² carbon network, where Anderson localisation may possibly play a major role.

1. Introduction

The intrinsic properties of graphene are strongly impacted by the addition of defects such as vacancies or adsorbed molecular species [1]. This alteration can be used to build very sensitive graphene-based sensors or to tune graphene electronic properties by defect engineering. In graphene, defect-induced change in physical properties is currently under intense investigation. Beyond single defects, a high level of disorder can be achieved with an electron beam [2] or ion beams. A high level of disorder induced by electron irradiation was shown to possibly result in the formation of a two-dimensional amorphous carbon lattice. Investigation of electronic transport in such 2D amorphous carbon was mostly restricted to theoretical work. The experimental characterisation of such disordered carbon network is highly desirable. Our motivation here is to generate a highly disordered carbon lattice and characterise its room temperature transport properties. We show that this can be realised using a helium ion microscope. Using He⁺ irradiation, we produced graphene sheets with controlled disorder and precise spatial position of the disorder. We measured our irradiated devices and show the existence of a transition into an insulating phase of two-terminal graphene nanodevices. This study will open possibilities for precision-engineered graphene nanoelectronic devices such as two-dimensional heterostructures and quantum dots, or the development of two-dimensional Anderson insulators.

2. Experimental

Our graphene is obtained from exfoliated graphite flakes which are transferred into a Si/SiO₂ substrate. The heavily doped Si is used as a gate electrode supporting a 300 nm-thick SiO₂ layer. The contact electrodes to the nanoribbons were made of Ti/Au and were defined using electron beam lithography and a lift-off process. Graphene nanoribbons were patterned using ebeam lithography and oxygen plasma etching. The nanoribbons were 1 um long and 200 nm wide. The devices were then transferred into a helium ion microscope (HIM) with an embedded pattern generator allowing patterning of structures as small as few nanometres beyond the capabilities of ebeam lithography, thanks to a beam spot size of 0.5 nm. In addition, the HIM offers the possibility of very precise He⁺ dose delivery at precise locations allowing deliberate introduction of point defects in the conduction channel. For the present study we irradiated the whole graphene channel in ultrahigh vacuum and measure its electrical characteristics for different irradiation doses ranging from 1 x 10¹⁵ to 6 x 10¹⁵ ion/cm².

3. Results and discussion

In figure 1 the Id-Vg characteristics are shown. In figure 1-a we show Id-Vg curves for non-irradiated and moderately irradiated graphene channel where the usual graphene characteristic is preserved showing an ambipolar behaviour and a neutrality point (NP). The conductivity varies linearly with the carrier concentration around the NP and shows a sub-linear behaviour far from the NP due to charged impurities scattering [3]. As expected a decrease in the drain current is observed as the dose is increased to 2 x 10¹⁵ ion/cm². Above 2 x 10¹⁵ ion/cm² the characteristics show a striking behaviour (figure 1-b): the devices no longer display the usual graphene Id-Vg curves but rather a flat characteristic on the electron side and a superlinear behaviour in the hole side.
Figure 1. a) Id-Vg curves for non-irradiated and moderately irradiated graphene nanoribbons. b) Id-Vg curves for heavily disordered graphene resulting from applied large doses. Here the drain voltage is Vd = 5 mV. Top image: highly irradiated graphene nanoribbons used in the current experiment. The false-colour areas are the metal contacts. The scale bar is 200 nm.

This behaviour becomes pronounced as the dose increases. A decrease in the drain current is also noticeable to a level of few tens of pA for largest dose of $6 \times 10^{15}$ ion/cm$^2$. We calculate the mobility $\mu$ from the transconductance:

$$g_m = \frac{\partial I_d}{\partial V_g} |_{V_d=\text{const}} = -\left(\frac{w}{L}\right)\cdot \mu \cdot c_g \cdot (V_g - V_{NP}) \quad (1)$$

Where $c_g$ is the capacitance per unit area, w and L are the width and the length of the channel.

Figure 2. Electron and hole mobility versus irradiation dose for several values of the gate voltage.

respectively, while $V_{NP}$ is the voltage at the neutrality point. In Figure 2 we show the electron and hole mobilities as a function of the irradiation dose for different values of the gate voltage. The mobility decreases abruptly after the third irradiation run which separates a conductive phase from an insulating phase. For instance, the electron mobility dropped by more than 5 orders of magnitude for the largest dose, from its value corresponding to non-irradiated channel.

Figure 3. TEM images of non-irradiated (a) and irradiated (b) CVD graphene showing different atomic arrangement for irradiated graphene. This image was taken at moderate dose. The scale bar is 2 nm.

To gain more insight into the observed behaviour, we examined the atomic structure of irradiated graphene using Transmission Electron Microscopy on non-irradiated and irradiated CVD graphene sheets. The result is shown in figure 3 where a clear difference in atomic arrangement is observed. Irradiated graphene loses its crystallinity and atoms rearrange themselves into more amorphous-like structure. This observation correlates well with our transport measurements which are also consistent with theoretical predictions where quantum interferences and Anderson localisation dominate the carrier transport in highly disordered graphene [4].

4. Conclusion

We conclude that irradiated graphene becomes an insulator above certain value of the irradiation dose. TEM images show that irradiate graphene becomes a highly disordered two-dimensional carbon network where Anderson localization may be responsible for the insulating behaviour.

References