Tuneable humidity gating of graphene's electronic properties

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Abstract

An important concern regarding graphene-based devices that are normally operated in ambient environments is that water and gas molecules reacting with graphene have an influence on devices performance and reliability. The presence of water is inevitable when graphene is exposed to air [1] and understanding its interaction with graphene is important in order to fully exploit possibilities for design and fabrication of graphene devices and sensing platforms.

Significant effort has been dedicated to both theoretical and experimental investigation of water on graphitic surfaces. However, in spite of these intense activities, a complete understanding of the water-graphene interaction is still lacking. From electronic properties point of view, water acts as a p-dopant, leading to a shift of the Fermi level in graphene and significantly affecting the electrical transport properties [2,3]. The information obtained from transport measurements is generalized over the entire device and is not correlated with the exact morphology of graphene, the presence of local adsorbates or structural defects.

In the current work, we employ scanning Kelvin probe microscopy (SKPM) to study the effect that water has on the electronic properties of epitaxial graphene, directly correlated with the local structural information, in an attempt to provide a systematic evaluation of the impact of ambient exposure on epitaxial graphene. We study the influence of relative humidity (RH=0-70%) changes on the surface potential of single-, bi- and tri-layer epitaxial graphene (1LG, 2LG and 3LG, respectively) and demonstrate the reversible process of water vapour adsorption and desorption on the various graphene domains.

A sequence of representative surface potential images of epitaxial graphene characterised by in-situ SKPM under various environments (ambient, vacuum and humid conditions) is displayed in Figure 1. In ambient conditions (Figure 1a), the surface potential map shows regions with two main distinct contrast levels: a bright one, given by two parallel stripes, associated with 2LG, superimposed on a dark contrast background of 1LG. This is furthermore highlighted by the corresponding histogram associated with the surface potential map (Figure 1e), displaying a bi-modal surface potential distribution. It is important to note that the topography (not shown here) acquired simultaneously with the surface potential image does not distinguish between the 1LG and 2LG and is mainly dominated by SiC parallel terraces with edges that promote the growth of subsequent layers.

Following ambient exposure, the sample was annealed at 150°C, under vacuum conditions (P = 6×10^{-6} mbar). Contrast inversion is observed in the associated surface potential image (Figure 1b) and the related histogram (Figure 1f), where 1LG now displays brighter contrast than 2LG. The contact potential difference, ΔV_{CPD}, measured between the 2LG and 1LG changes sign compared to ambient and is consistent with lower carrier (electron) concentration in 2LG compared to 1LG in vacuum, $n_e(2LG)<n_e(1LG)$. The opposite is observed in ambient, i.e. higher carrier concentration for 2LG in comparison to 1LG, $n_e(2LG)>n_e(1LG)$. The observed effect is explained in terms of an increase of the electron concentration in both 1LG and 2LG on ambient-vacuum transition due to desorption of environmental p-dopants. V_{CPD} values of 1LG and 2LG reproducibly restored to initial values whenever re-exposing the sample to ambient, indicating that the properties of graphene are strongly influenced by surface charge imposed by atmospheric adsorbates.

In the next set of experiments following vacuum annealing, the sample was exposed first to dry nitrogen at atmospheric pressure and then to varying humidity levels, ranging between 10% and 70%. Only the images acquired under the lowest (10%) and the highest (70%) humidity atmospheres are shown in Figure 1c and Figure 1d, respectively. The initially negative value of ΔV_{CPD} between 2LG and 1LG of the vacuum annealed sample gradually decreases in absolute value with increasing humidity, passes through 0 at ~ 50% humidity and reaches a positive value for 70% humidity levels. As shown in Figure 1d, 2LG inverts contrast at 70% humidity, although not reaching the level observed in ambient, indicating that other factors in addition to water can significantly affect the surface potential and the electronic properties of graphene.

The above observations will be presented, discussing in detail the charge transfer at graphene-water interface, as well as doping levels of 1LG compared to those of 2LG and 3LG upon water

adsorption/desorption, taking into account the interplay between carrier concentrations in graphene, the underlying substrate and atmospheric adsorbates.

The results demonstrate the importance of surface studies of graphene in ambient, as well as at elevated temperature and humidity conditions in order to fully understand the underlying physical phenomena and control the electronic properties of proposed graphene devices.

References

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Figures
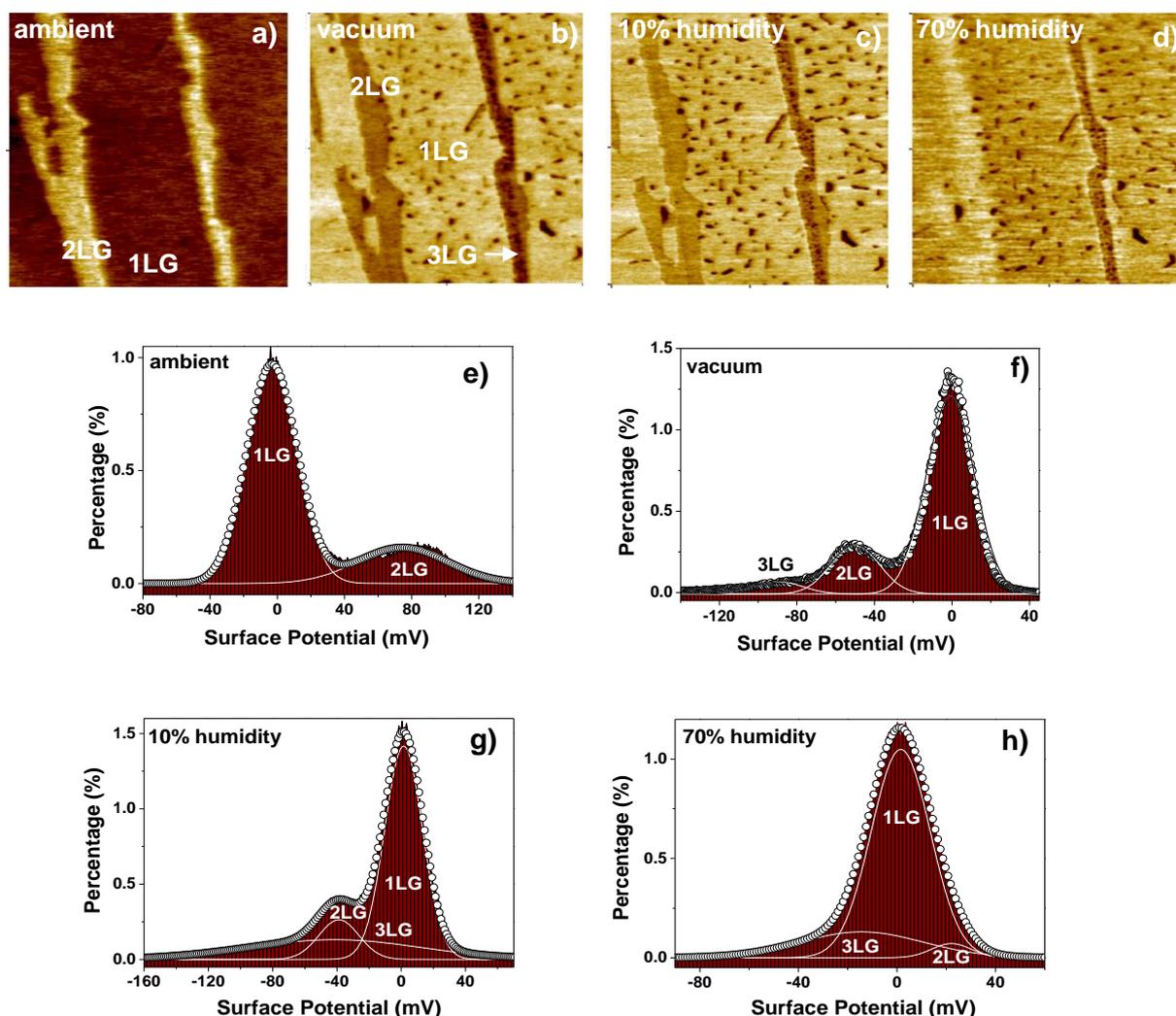
Figure 1: Sequence of surface potential images collected on the same region of the graphene sample, while environmental conditions were changing in the following order: ambient (a), vacuum after annealing at 150°C (b), 10% humidity (c), 70% humidity (d). The scan size is (10x10) μm^2 for all images. (e)-(h) Histograms showing relative V_{CPD} values between individual layers correspond to images (a-d), respectively.