Bandgap opening in hydrogenated graphene studied by electronic transport and STM

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Most electronic logic applications require material with a bandgap. Although graphene is a gapless conductor a gap can be created by chemical modification, e.g. hydrogenation [1], in which adsorbed H changes the orbital hybridization of carbon from sp2 to sp3. Depending on the degree of hydrogenation one can tune the graphene's transport properties from zero-gap semiconductor to insulator. In experiment we hydrogenate graphene by RF plasma with a gas mixture of Ar/H_2 (85:15). Here, the introduced defects like hydrogen adsorbates or argon induced vacancies, can be characterized by the ratio of the D and G band in Raman spectrum [2]. We present the systematic studies of electronic transport in graphene depending on plasma exposure time and report the influence of the amount of defects on graphene carrier mobilities and mean free path.

Additionally, we have explored the change in the electronic properties of CVD graphene and graphite after hydrogenation by scanning tunneling microscopy (STM). STM topography images reveal a non-uniform hydrogen chemisorption on the surface, forming islands, which cover roughly 20% of the surface. From the statistical analysis of scanning tunneling spectroscopy (STS) traces we have observed a hydrogen induced bandgap opening, with an average value of 0.6 eV. Further, we have studied the electronic and structural changes of the samples after a moderate thermal annealing which should de-hydrogenate the surface. Remarkably, it has been found that even though topographic changes are not fully reversible after annealing, the bandgap is closed. The full cycle of hydrogenation/de-hydrogenation can be repeated with similar results.

References

[1] Elias, D.C. et al. *Science (2009), 323*, 610-613
[2] Lucchese, M.M. et al. *Carbon 48 (2010)*, 1592-1597

Figures

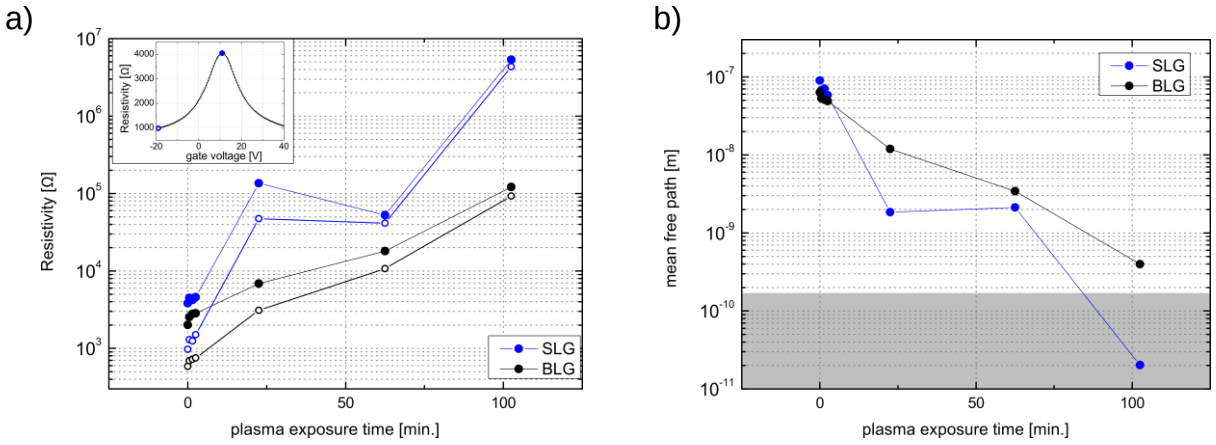

Figure 1. (a) Resistivity of single (blue) and double layer graphene (black) after several exposures to hydrogen plasma. Filled circles represent the resistivity at the Dirac point, open circles represent the resistivity in metallic regime (at $2 \cdot 10^{12}$ cm^{-2} carrier density). The inset present the single resistivity curve for SLG. (b) Mean free path of charge carriers in graphene after the exposures. The gray area indicates the values below the length of C-C bond, where the calculations of the mean free path are no longer valid.

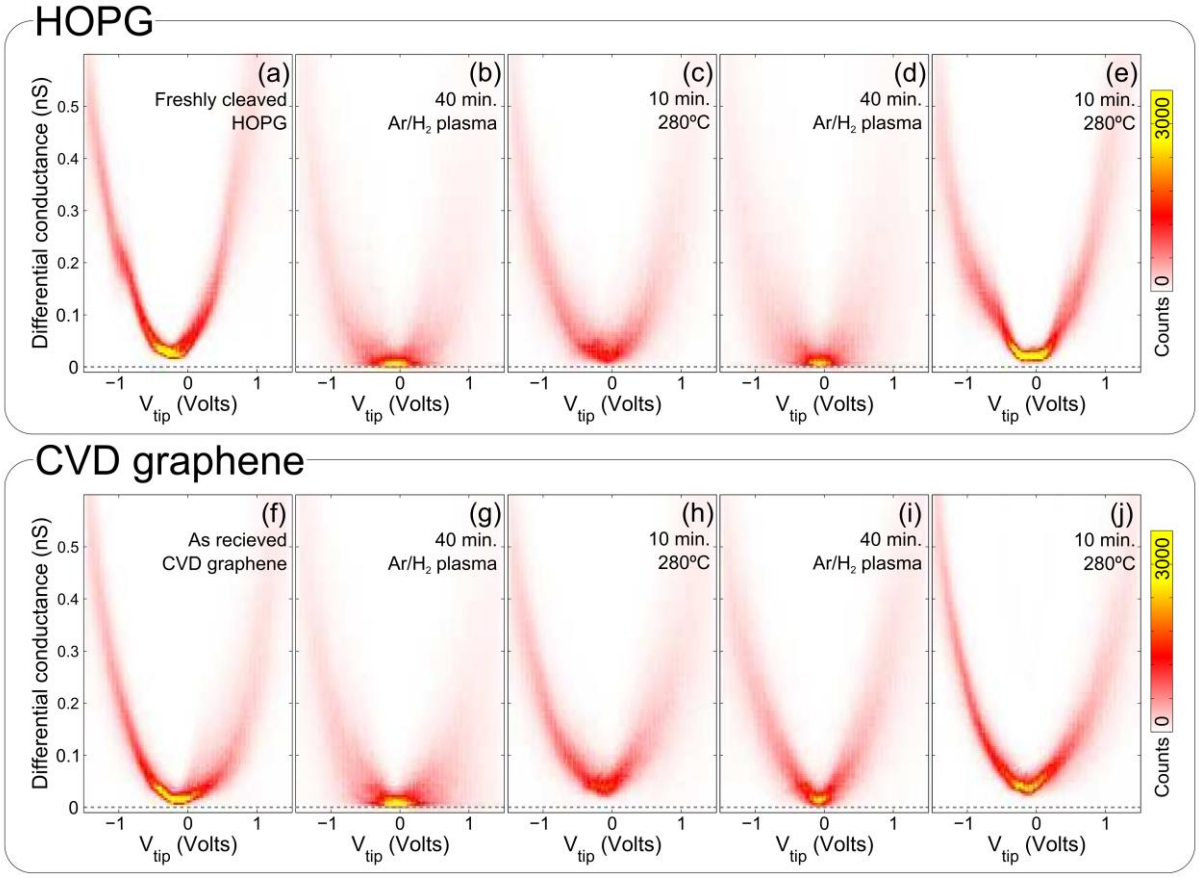

Figure 2. A set of 2D histogram of STM spectroscopic dI/dV traces acquired for two different samples: HOPG graphite and CVD graphene on Ni at different processing stages. In each histogram 2000 traces are collected at random STM tip locations on the sample. Initially both samples show metallic behaviour (dI/dV>0) (a, f), while after hydrogenation (b,g) the semiconducting behaviour, with the bandgap above 0.6 eV is present. Annealing recovers the metallic behaviour (c,h). The procedure of hydrogenation and annealing can be repeated (d,e,i,j)leading to similar properties.