

Hydrogenation and etching of a graphene during exposure to hydrogen radicals

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One-side hydrogenation of a graphene monolayer (1LG) on SiO₂ substrate is of great interest for microelectronic devices since it can open the graphene energy band gap. Experimental evidence of hydrogenated 1LG/SiO₂ is scarce and often lacks characterization at the nanoscale. The chemisorption of hydrogen radicals is not easily achieved, as this reaction may compete with the release of H₂ through the Eley-Rideal mechanism [1] or the removal of carbon by etching. We investigated the one-side hydrogenation of exfoliated 1LG flake transferred onto SiO₂/Si substrate, and then exposed to a flow of H radicals using the hydrogenation process assisted by hot-filament [2] at two different exposure times. We discuss a regime where hole formation coexists with hydrogen chemisorption. From thermally induced dehydrogenation experiments followed by Raman spectroscopy analysis on the one hand, and electronic band structure density functional theory (DFT) calculations as a function of hydrogenation rate on the other hand, hydrogenation coverage is estimated to be below 1 at. % (figure 1). The optical contrast is shown to decrease upon hydrogenation, which is due to both the presence of holes (revealed by atomic force microscopy (AFM) images) (figure 1) and the loss of the graphene Dirac cones. On the most exposed samples, it is possible to recover the Raman signal by increasing the power of the laser used. We discuss mechanisms for the creation of holes by using DFT calculations and two structure models based on isolated hydrogenated carbon clusters. Stress alone, cannot be at the origin of the carbon-carbon bond breaking needed to create holes. Another molecular process is required. This research is supported by: (i) ANR-21-CE09-0003 GLADIATOR project and NanoX ANR-17-EURE-0009 (France), (ii) CNRS through the French-Dominican International Research Project NEWCA (France), (iii) FONDOCYT, grant No. 2020-2021-1A1-066 and No. 2022-1A1-095 (Dominican Republic) and (iv) ONRG and SOARD grant No. N62909-24-1-2000.

Reference

- [1] D. Eley, E. Rideal, Nature 146 (1940) 401.
- [2] T. Fournier, K. Cruz, M. Monthieux et al., Mat. Chem Phys. 321 (2024) 129490.

Figure

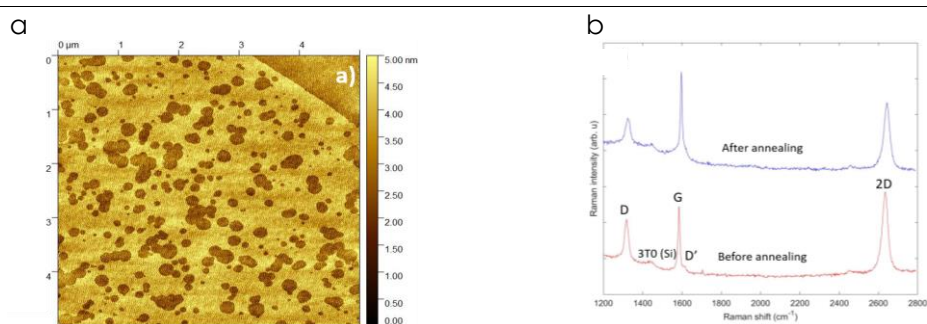


Figure 1: (a) Topographic AFM image of 1LG exposed to H radicals during 10-minute, showing holes (dark contrast). (b) Raman spectra corresponding to a, before (red) and after (blue) annealing under Ar/H₂ atmosphere (95/5 vol.%) at 400°C for 1 hour.