HYDROGENATION OF GRAPHENES FROM H RADICALS

Pascal Puech,[1] Fabrice Piazza,[2] Marc Monthioux,[1] Iann Gerber,[3] Benjamin Lassagne,[3] Lionel Petit,[3] and Tom Fournier[3]

[1]CEMES, UPR-8011, CNRS, Université de Toulouse, Toulouse, France
[2]Laboratorio de Nanociencia, PUCMM, Santiago de Los Caballeros, Dominican Republic
[3]LPCNO, UMR 5215, CNRS, INSA, Université de Toulouse, Toulouse, France

The hydrogenation of single layer graphene (1LG) is controversial as its confirmation is difficult to obtain, and many reports claim for the related sp^2-C to sp^3-C transformation without clear evidence or based on inconsistent observations [1].

Here, a simple but very efficient hydrogenation route was used. The decomposition of H_2 at high temperature on tungsten heating wires was carried out to deposit H radicals on either bilayer graphene (2LG) deposited on a transmission electron microscopy (TEM) Cu grid or various graphenes (from 1LG to few-layer graphene (FLG), exfoliated from HOPG) deposited on an Si-based interferential substrate as reported on figure 1.

TEM characterization of such materials is limited because hydrogenated layers are not stable under the energy range of the electron beams currently used (typically, > 50 keV). Consistently, very low energy electron diffraction (5 keV) was able to discriminate between hydrogenated and non-hydrogenated graphene, and between various stacking sequences as well [2]. Based on this method, a new approach able to take into account misorientations is proposed, which makes the analysis of the diffraction patterns accessible even if the material is affected by local distortions. We will present these results in detail, before (see figure 2) and after hydrogenation.

On the other hand, multi-wavelength Raman spectroscopy succeeded in demonstrating the actual conversion of 2LG into diamane [3], thanks to the sensitivity of visible light excitation to C(sp^2) domains and that of UV light excitation to C(sp^3) domains. It is complementary to Infrared microscopy which is able to reveal C(sp^3)-H but remains qualitative only as the conversion rate is not accessible. The interferential substrate was useful to optically reveal the hydrogenation-promoted C(sp^2)-to-C(sp^3) conversion by a mere color change while enabling Raman spectroscopy. Whereas obtaining the signal of both D and G bands after hydrogenation is a clear signature of partial conversion only, we show that 1LG exposed to hydrogen radicals becomes transparent in the visible light range without exhibiting any Raman signal. We also discuss the case of the partial hydrogenation of 2LG (either AB stacked or twisted considering thermodynamics considerations proposed by Erohin et al [4].

References

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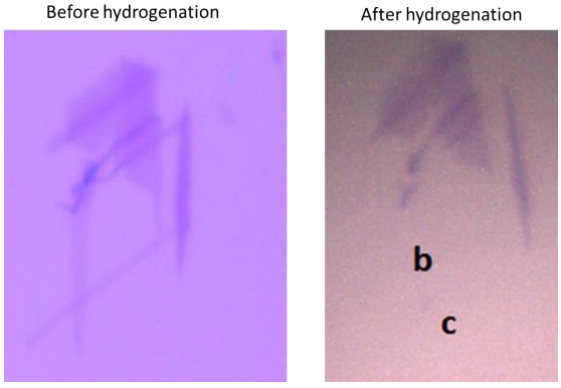

Figure 1: Contrast change after hydrogenation. (b) 1LG, (c) twisted 2LG.

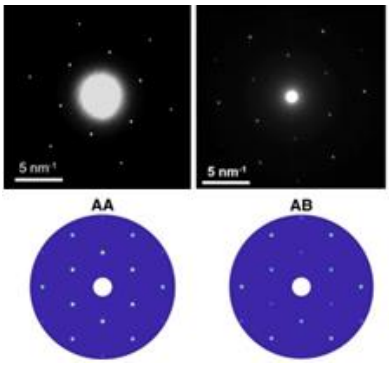

Figure 2: low voltage (5 keV) diffraction patterns for 2LG with AA and AB stacking. Top: experimental; bottom: calculated.

corresponding author : pascal.puech@cemes.fr