Revisiting EELS Characterization and its Coupling with Raman Spectroscopy: Chemical Inhomogeneities at the Nanoscale of DLC and Nanodiamond Thin Films

L. Lajaunie[1] C. Pardanaud[2] J. G. Buijnsters[3] R. Arenal[1,4]

[1]Laboratorio de Microscopias Avanzadas, Instituto de Nanociencia de Aragon, Universidad de Zaragoza, 50018 Zaragoza, Spain
[2]Aix-Marseille Université-CNRS, PIIM, 13397 Marseille cedex 20, France
[3]Department of Precision and Microsystems Engineering, Delft University of Technology, Mekelweg 2, 2628 CD Delft, The Netherlands
[4]ARAID Foundation, 50018 Zaragoza, Spain

The long-term stability of the properties of hydrogenated amorphous carbon (a:C-H) thin films makes them very promising materials for numerous applications, including coatings for spatial applications [1]. For improving their performances, a full understanding of their local chemistry is highly required. Fifteen years ago, according to the seminal work of Ferrari et al., [2] EELS was the most used technique to get such kind of quantitative information on these materials. Nowadays, the complexity of the physics phenomena behind EELS is well known [3], but this technique is regarded as time-consuming and difficult to interpret properly. Other optical techniques, such as Raman spectroscopy, are now clearly favored by the scientific community. However, these macroscopic techniques still lack the high spatial resolution. This limitation can be overcome by STEM-EELS, which offers the possibility of getting direct chemical information at the local (atomic) scale.

In this contribution, we will revisit the procedures to extract proper and reliably quantitative chemical information from EELS spectra. In addition, the coupling of multi-wavelength Raman and EELS spectroscopies to obtain a wealth of chemical information will be discussed. Our results provide a complete combination of C-hybridization, spatial elemental analyses and structural defects studies for shedding light on these complex materials. In particular, we will show how the deposition process induces a gradient of sp^2 ratio in the thin films and how this gradient is modified as a function of the annealing time [4]. In addition, recent results on nanocrystalline and microcrystalline CVD diamond films will be presented [5]. Surprisingly, strong in-depth inhomogeneities of the local chemistry has been highlighted and the evolution of the morphology, nanostructure and composition of the films, as a function of methane in excess of hydrogen, will be discussed.

Acknowledgement: We acknowledge funding from the EU under Grant Agreement 312483-ESTEEM2, Grant Agreement 696656 Graphene Flagship and the Spanish Ministerio de Economia y Competitividad (FIS2013-46159-C3-3-P).

[1] A. Rusanov et al., *Carbon* **81**, 788-799 (2015).
[2] A.C. Ferrari et al., *Phys. Rev. B* **62 (16)**, 11089 (2000).
[3] P. Schattschneider et al., *Phys. Rev. B* **72**, 045142 (2005).
[4] L. Lajaunie, C. Pardanaud, C. Martin, P. Puech, C. Hu, M.J. Biggs and R. Arenal, *Carbon* **112**, 149-161 (2017).
[5] J.G. Buijnsters, C. Pardanaud, L. Vazquez, R. Gago, J.L. Endrino, L. Lajaunie and R. Arenal, *In Preparation* NT (2017).