Revisiting EELS investigations and its coupling with Raman spectroscopy: chemical inhomogeneities at the nanoscale of hydrogenated amorphous carbon thin films

Arenal, R.[1], Lajaunie, L.[1] and Pardanaud, C.[2]

[1] Universidad de Zaragoza, Spain, [2] Aix-Marseille Université-CNRS, France

The long-term stability of the properties of hydrogenated amorphous carbon (a:C-H) thin films makes them very promising materials for numerous applications [1]. For improving their performances, a full understanding of their local chemistry is highly required. The seminal work of Ferrari et al. [2] showed that EELS was the most appropriated 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 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, see Fig. 1 [4].

[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]. We acknowledge funding from the EU under Grant Agreement 696656 Graphene Flagship and the Spanish Ministerio de Economia y Competitividad (MAT2016-79776-P (AEI/ 577 FEDER, UE)).

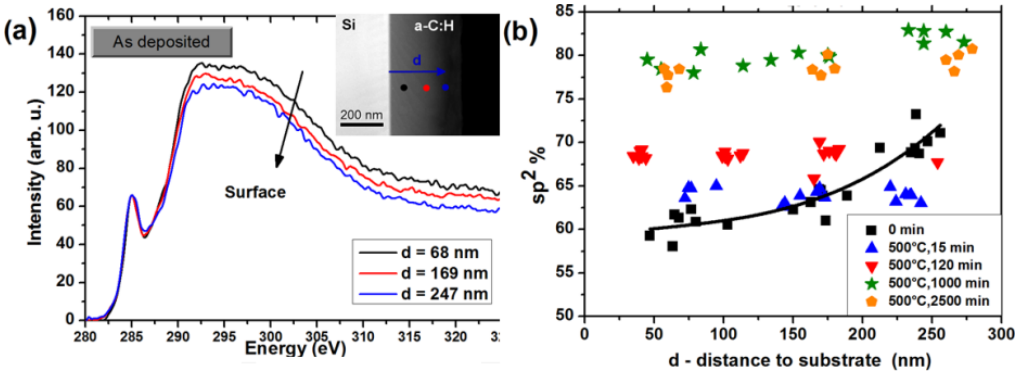

Fig. 1 (a) Variation of the C-K ELNES of the as-deposited sample with the distance to the substrate. The inset shows a low-magnification STEM-HAADF micrograph of the as-deposited sample highlighting how the d parameter (distance between the electron probe and the substrate) is defined. **(b)** Variation of the sp^2 fraction, determined by EELS, as a function of the distance between the electron probe and the substrate. The black line is a guide for the eyes.