Understanding electronic interactions at the graphene-electrolyte interface

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Due to the unique electronic and chemical properties of graphene, electronic devices based on this material are nowadays widely used in a number of applications; specially, in the field of biosensing, in which graphene is in direct contact with an aqueous electrolyte, which contains ions and molecules of different nature. The previous works concerning 2D materials-based sensors reveal extremely large dispersions concerning graphene's sensitivity to specific analytes. Thus, understanding the graphene-electrolyte interface is crucial, also taking into account the influence that the adsorbed surface charge and the substrate may have on it. To this end, in this work, spectroscopic techniques such as Raman and impedance were used. We demonstrate that Raman enables addressing charge carriers in graphene supported on substrates of different nature, conductive or insulating; as well as being an indirect measurement of the adsorbed charge in graphene [1]. Complementary, from impedance spectroscopy we obtain information about the capacitance dependence with the parameters of study: pH, ionic concentration, substrate nature and applied voltage [2]. Additionally, theoretical simulations help in understanding the effect of involved phenomena like the nature of the adsorbed charge and the coupling between graphene and the substrate. Further characterization tools, including XPS, AFM and KPFM, are used to complete this study. Our results reveal the intriguing mechanisms of charge modulation in graphene via the electrolyte, and shed some light onto the intrinsic sensing capabilities of graphene (undoped and free of defects) and their modulation through the adsorbed surface charge and/or the substrate electronic properties, which is very precious information for numerous applications involving graphene-liquid interfaces.

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References

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[2] J Xia, F Chen, J Li, Nongjian T, Nat Nanotechnol. 4 (2009) 505.

Figures

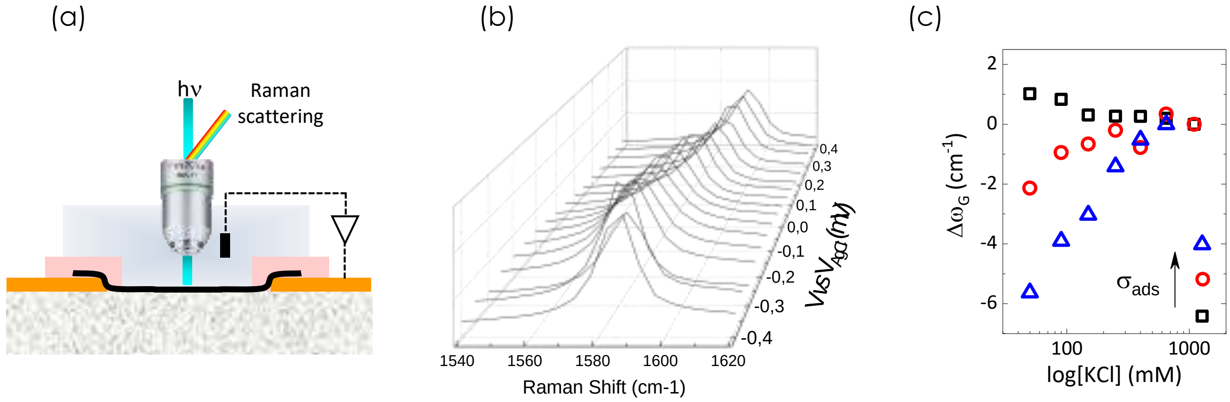

Figure 1. Schematic lateral view of the graphene electrode device for in situ Raman characterization. Raman spectra as a function of the applied voltage. Modulation of the Fermi level of graphene through the electrolyte ionic strength for adsorbed charge concentration.