Development of *ab-initio* method to multiwave-length dependent Raman spectroscopy of 2D-nanomaterials

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Abstract

We developed how to analyse non-resonant Raman spectra by using the Placzek approximation based on semi-classical approximation of electronic and vibrational transition energies [1]. Upon calculated frequency-dependent dielectric tensors, we can compute polarized Raman intensities by averaging over all in- and out-plane polarizations, allowing us to capture Raman resonance effects in first-order Raman scattering. Our *first-principle* investigation covered a broad family of nanocarbons from well-known cases, e.g. graphene, fullerene, and graphene nanoribbons till systems with unique architecture, like fully flattened single-walled carbon nanotubes and graphitic prismatic edge dispersion of the D peak, attributed to the curvature which acts as activating defect as confirmed in recent experiments [2,3]. Given its robustness, the application of our approach can be extended to reproduce the Raman characterisation of other nanomaterials beyond graphene, like phosphorus allotropes encapsulated in single-walled carbon nanotubes [4].

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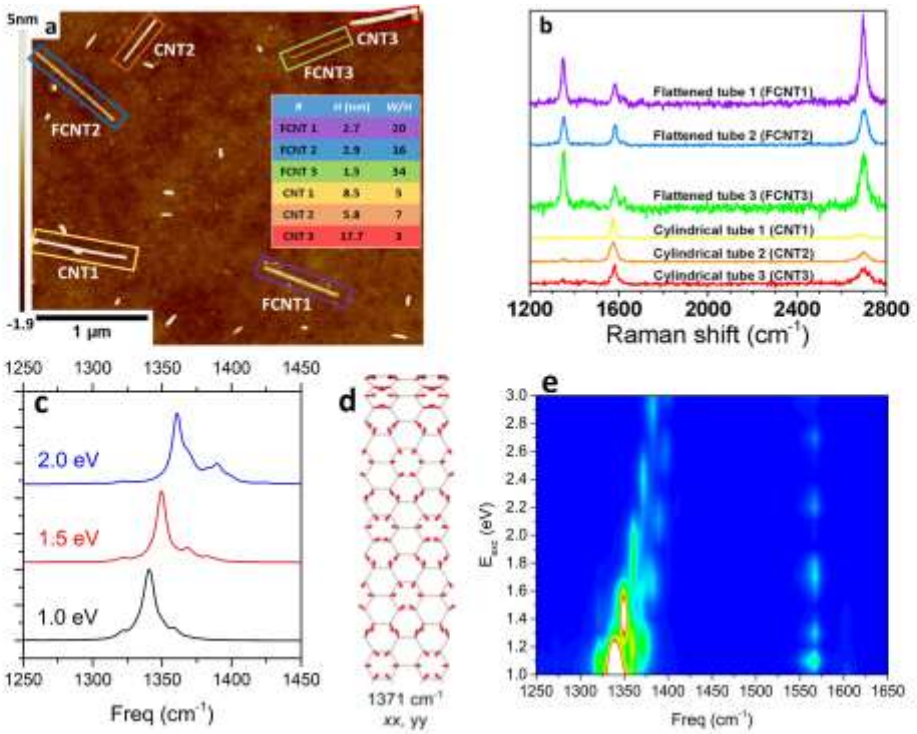

Figure 1: (a) Atomic Force Microscopy height image of 3 flattened (FCNT) and 3 cylindrical (CNT). (b) Corresponding Raman spectra normalized over the G peak recorded at 2.4 eV. (c) Calculated Raman spectra of a zig-zag (70,0) fully FCNT at different excitation energies. (d) Eigendisplacements of the strongest vibrations contributing to the Raman D peak with its non-zero Raman tensor components. (e) Colour mapping representation of dispersive character of the D peak.