Uniaxial tension of 2D membranes such as graphene; is orthogonal buckling avoidable?

I. Polyzos[1], E. Koukaras[1], D. Sfyris[1], G. Tsoukleri[1], M. G. Pastore Carbone [1], J. Parthenios[1], K. Papagelis[2,] and C. Galiotis[3*]*

[1]Foundation of Research and Technology Hellas, Institute of Chemical Engineering and High Temperature Processes,, GR-26504 Patras (Greece)
[2]Department of Materials Science, University of Patras, GR-26504 Patras (Greece)
[3*] Foundation of Research and Technology Hellas, Institute of Chemical Engineering and High Temperature Processe4, GR-26504 Patras (Greece) , Department of Chemical Engineering, University of Patras, GR-26504 Patras (Greece), e-mail: ipolyzos@iceht.forth.gr; c.galiotis@iceht.forth.gr, galiotis@chemeng.upatras.gr

Graphene is an ideal 2D crystal which is believed to possess a unique combination of mechanical properties in tension; that is high stiffness (~1 TPa), high strength (>100 GPa) but also high ductility (>20%) [1]. Several experiments confirm graphene as the strongest material ever measured but there are still a number of open questions as the above values have been derived from a bending experiment employing certain assumptions regarding the bending stiffness of the flake and the expected non-linear form of the derived stress-strain curve. Since the flake should be much weaker in compression there is uncertainty as to whether the bending experiments performed by Lee et al[1]were also accompanied by lateral wrinkling of the membrane. Thus, there is a real need for a purely axial tensile experiment without the presence of lateral (orthogonal) wrinkles so as to assess true tensile stress-strain characteristics of graphene.In general it is expected and already verified experimentally, that a thin film can withstand relatively large tensile strains in air without early fracture, whereas in compression monolayer graphene is expected to buckleat extremely low strains[2]. Furthermore, when an exfoliated monolayer graphene is stretched axially in one direction should always be accompanied by the formation of lateral (orthogonal) wrinkles or buckles which render pure axial experiments untenable, phenomenon which should be prevalent for any future 2D materials, has not as yet been fully studied, predicted or, even, exploited[3].

In the current work e-beam lithography was used to suspend a section of a single layer graphene flake previously embedded in PMMA. An intrinsic gradient of uniaxial strain is applied to graphene through the fabrication procedure. Raman spectroscopy, a fast and non-invasive optical probing technique, was employed to measure the actual values of strain with high frequency and spatial. A range of axial strains up to ~0.8% was applied to suspended graphene. For the first time in the literature, both the well-known G and 2D peaks of graphene were clearly splitted and shifted considerably with strain. Shift rates were similar to those predicted theoretically, unequivocally proving -based on fundamental physics- that the material is axially stressed. A systematic undulation of the wavenumber and intensity in the transverse direction of strain axis is clearly observed (Fig.1). This behavior can be explained by the formation of a buckling wave in the transverse direction; the values near the crest and trough of the buckling wave are affected by the corresponding transverse tensile and compressive components. The wrinkle formation causes a variation of local axial strain of the order of 0.05 to 0.22%.

Moreover, recently, we have examined the mechanical behavior of several monolayer graphene flakes with various length to width ratio for small deformations, collecting simultaneously Raman spectra[2, 4, 5]. Under compression the critical strain to buckling was found to be ~–0.6% and independent of the flake's dimensions. Also, it was found that a minimum length of about 4 μm is needed in order to have sufficient stress transfer from the surrounded polymer to the graphene[5]. Thus, we exploit the above findings by altering the geometry of the flakes, and design graphene strips (micro-ribbons) of specific dimensions which when embedded to thermoplastic and thermosetting polymer matrices can be stretched to undergo large deformation without simultaneous buckling in the other direction. The dimensions of the micro-ribbons range between 1-5 μm (width) and 10-50 μm (length) while the ratio length over width is kept constant and equal to 10/1.

Finally, in addition to the above experiments, we have performed theoretical calculations to gain insight on aspects that may affect, induce or inhibit the formulation of orthogonal buckling. The theoretical calculations consist of molecular dynamics simulations the results of which are processed and utilized in mathematical models. The initial goal is to identify effects that arise by partial clamping at the edges of nanographenes, record any systematic behavior that may be identified and formulate a mathematical basis for the description when mixed boundary conditions are imposed. Several series of simulations have been performed changing defining parameters such as the percentage of the edges that is clamped on which the tensile strain is applied, the aspect ratio of the flakes, and the size of the flakes for each given aspect ratio. This provides a basis for a more realistic description of the real graphenes through edges with sequential regions of clamping and free boundaries, instead of idealized fully clamped or fully free edges. The results clearly show that orthogonal buckling is readily manifested when the edges of the nanographenes are partially clamped. Furthermore, the buckling is systematically quenched in all cases upon increase of the percentage of the edges that is clamped. The prospect is to identify the dependence of the amplitudes and the transverse wavelengths on the clamping conditions and the aspect ratios of the flakes.

References

[1] C. Lee et al., Science **321**, (2008), 385.

[2] O. Frank et al., ACS Nano **4**, (2010), 3131.

[3] J. C. Meyer et al., Nature **446**, (2007), 60.

[4] G. Tsoukleri et al., Small **5**, (2009), 2397.

[5] C. Androulidakis et al., Sci. Rep. **4**(2014), 5271.

[6] This research was supported by: i) the Graphene FET Flagship (‘‘Graphene-Based Revolutions in ICT And Beyond’’- Grant agreement no: 604391).”: ii) European Union (European Social Fund-ESF) and Greek national funds through the Operational Program ‘‘Education and Lifelong Learning’’ of the National Strategic Reference Framework (NSRF) - Research Funding Program: ERC-10 ‘‘Deformation, Yield and Failure of Graphene and Graphene-based Nanocomposites’’, iii) the European Research Council (ERC Advanced Grant 2013) no. 321124, “Tailor Graphene” and iv) “Dr. Polyzos would like to acknowledge the support of the action “Supporting Postdoctoral Researchers” of the Operational Program "Education and Lifelong Learning", which is co-financed by the European Social Fund (ESF) and the Greek State through the General Secretariat for Research and Technology.

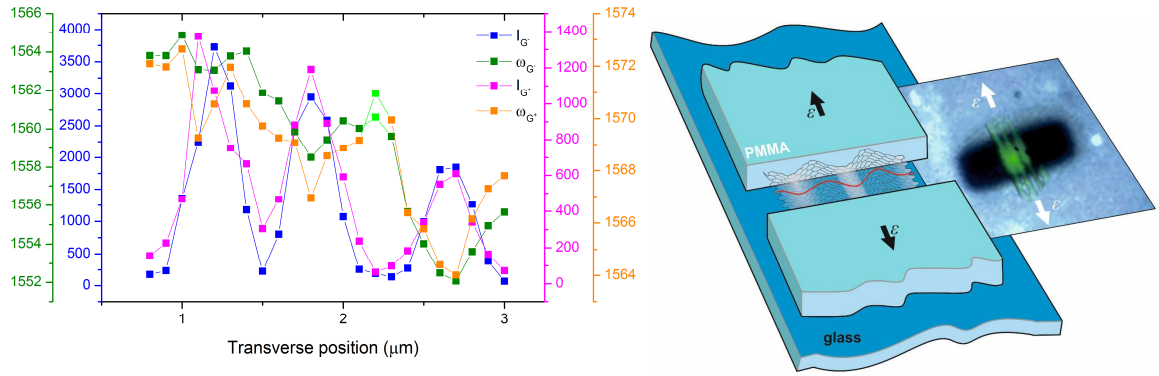

Fig.1 (a)Raman intensity (I) and frequency (ω) variation of the G and 2D sub-peaks in the transverse direction to strain axis.(b) Schematic of wrinkle (buckle) formation due to lateral compression and Raman map of the 2D peak intensity of graphene flake, I(ω_{2D}).