

Efficient mechanical loading of few layer graphene flakes: experiment and modeling

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Abstract

Recently, there has been a growing interest in trilayer and few layer graphene materials because of their interesting properties. In these systems the electronic, optical and vibrational properties are distinct from those of single-layer graphene and strongly depended on the crystallographic stacking of the individual graphene sheets providing an alternative degree of freedom to tune graphene properties [1]. Trilayers exhibit two main stacking order configurations, namely the energetically more stable Bernal (or ABA) and the rhombohedral (or ABC) stacking [2]. The ABA stacked trilayers are semimetals showing overlapping linear and quadratic electronic dispersion near the Fermi level with an electrically tunable band overlap, while ABC stacked trilayers are predicted to be semiconductors exhibiting cubic dispersion with a tunable band gap similar to that of bilayer graphene.

Raman spectroscopy has been proven a very successful technique to investigate the effect of mechanical deformation on graphene materials under uniaxial tension and compression [3, 4, 5] or hydrostatic pressure [6]. Therefore, monitoring optical phonons it seems the clearest and simplest way to quantify the macroscopic stress/strain imparted to graphene sheets.

In this work, we have been subjecting to tension various single-, bi-, tri- and few-layer graphene samples, embedded into the upper surface of a PMMA cantilever and covered by a ~100nm thickness polymeric film, while their Raman spectrum is recorded simultaneously. The mechanical response is monitored by the shift of the G and 2D Raman lines with strain, using the 785nm (1.58 eV) excitation wavelength.

The investigated samples exhibit a notable peculiarity where each graphene unit does not form a stack having ideally sharp edges in the atomic scale. The single layer graphene (1LG) is in direct contact with the polymer substrate which is the footprint of the whole unit. The other layers forming the bilayer (2LG), trilayer (3LG) and few layer graphene (FLG) are extended above the monolayer partially overlapping in a ladder-like fashion (figure 1).

We have observed for the first time the strain induced lifting of the degeneracy of the E_{2g} mode in 3LG and FLG. The shift rates are similar to those observed for single and bilayer graphene [5]. The only difference is an up-shift of the onset of splitting due to the full width of half maximum enhancement with the number of layers. This behaviour indicates an efficient stress transfer across the graphene-polymer interface.

On the other hand, for 3LG and FLG a linear dependence between the 2D Raman frequency and strain is observed. The 2D strain sensitivity in both cases is about 55 $\text{cm}^{-1}/\%$. The results are different from those of Gong et al [7] where they observed smaller 2D band shift rates (47 $\text{cm}^{-1}/\%$ for 3LG and 40 $\text{cm}^{-1}/\%$ for FLG) attributed to poor stress transfer efficiency upon increasing the number of layers. The measured slopes of the studied samples show negligible dependence on the number of graphene layers as a consequence of the morphology of the studied sample where each internal graphene layer can be uniaxially loaded due to the direct contact with the polymer molecules.

Molecular dynamics simulations using an improved long-range reactive bond-order potential for carbon (LCBOP) [8] with the long-range interactions cutoff at 0.6 nm to ensure interplanar binding in graphite have shown that for 3LG the middle layer can sustain tensile strains only up to 0.06% external strain before sliding. Besides, if 20% of the middle layer edge atoms located on both sides perpendicular to the uniaxial strain axis experience the applied force then the middle layer follow the induced deformation with efficiency of almost 85%. Therefore, in the case of embedded graphene flakes where the inner layers protruding from the upper and lower ones the interaction of the uncovered parts of the flake with the polymer matrix can enhance dramatically the load transfer from the matrix to the inclusions. The observed behavior have important implications in the level of reinforcement in polymer composites where we can engineer the degree of reinforcing efficiency for few layer graphene or even nano-graphite by suitably shaping the edges of the inner graphene layers.

References

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Figures

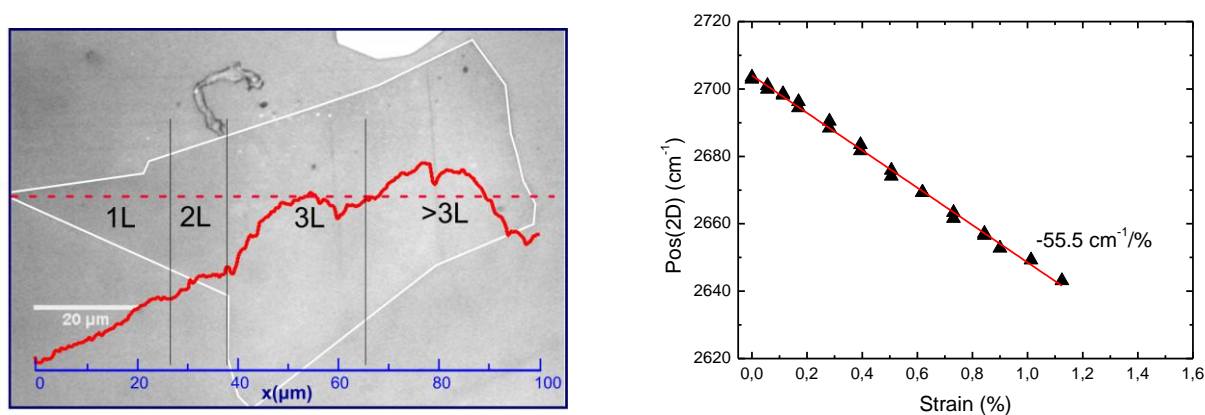


Figure 1. (Left panel) Morphology of the investigated flake. (Right panel) 2D band frequency dependence as a function of uniaxial strain for FLG.