Understanding the Potential of Graphene within Composites: Interfacial Stress Transfer in Ideal Graphene Composites

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Graphene is one of the stiffest and strongest known materials with a Young's modulus of the order of 1 TPa and a fracture stress ~ 130 GPa. These properties make graphene an ideal candidate for use as a reinforcement in high-performance composites. However, being one-atom thick crystalline material, graphene poses several rather fundamental questions: (1) Can decades of research on carbon based composites be applied to such ultimately thin crystalline material? (2) Would traditionally used continuous mechanics still be valid at atomic level? (3) How does the macroscopic matrix interact with microscopic graphene crystals (in terms of stress transfer, for instance) and what kind of theoretical description would be appropriate? (4) How does stress transfer between the layers in bilayer and thicker graphene?

We have prepared model composites consisting of single graphene flakes (monolayer and thicker) sandwiched between thin polymer films and employed Raman spectroscopy to monitor stress transfer from the polymer matrix to the graphene during deformation of the composite [1]. Typically, a PMMA beam was coated with 300 nm of epoxy, onto which mechanically exfoliated graphene was placed, finally a 50 nm PMMA coating was then spun onto the graphene. The coated beam was tested in 4-point bending and the Raman spectra were collected to confirm the morphology of the flake being studied and map the strain within it.

The rate of peak shift of the G' peak with applied strain has been shown for carbon reinforcements to be proportional to the effective modulus of the filler in the composite [2]. (This effective modulus takes into account the efficiency of the polymer-filler interface). A shift of ~ 35 cm^{-1}/% was obtained for the monolayer graphene upon initial loading, corresponding to an modulus of ~ 0.6 TPa. The polymer-graphene interface was found to fail at high strains but would heal upon relaxation of the sample. The peak shift rate upon unloading, however, was 60 cm^{-1}/%. This shift rate is the highest we have ever observed in any composite system and corresponds to an effective modulus of ~ 1 TPa. This result shows that the full modulus of graphene can be achieved within a polymer matrix.

Mapping the G' peak shift across the flake showed that the strain in a flake was virtually zero at the ends of the flake and built up to the applied, global strain value in the middle. This behaviour was successfully modeled using the continuum shear-lag theory, which is the established model for macroscale fillers. In particular, the diamond shape of the flake that was studied allowed an accurate fit of the model since it meant that the strain data over a range of aspect ratios could be obtained. The maximum shear force at the polymer-graphene interface was calculated as 3 MPa, which is an order magnitude less than that typically achieved in conventional carbon fibre-epoxy composites. Such a low shear force maybe expected, given the weak nature of the van der Waal forces at the interface. One result of these poor interfacial properties is that the critical length of the graphene required to obtain efficient reinforcement was found to be 30 microns. This result implies that in order for graphene to be used in structural composites, the graphene flakes either need to be large (> 30 microns long) or chemically functionalised to improve the interfacial strength with the matrix.

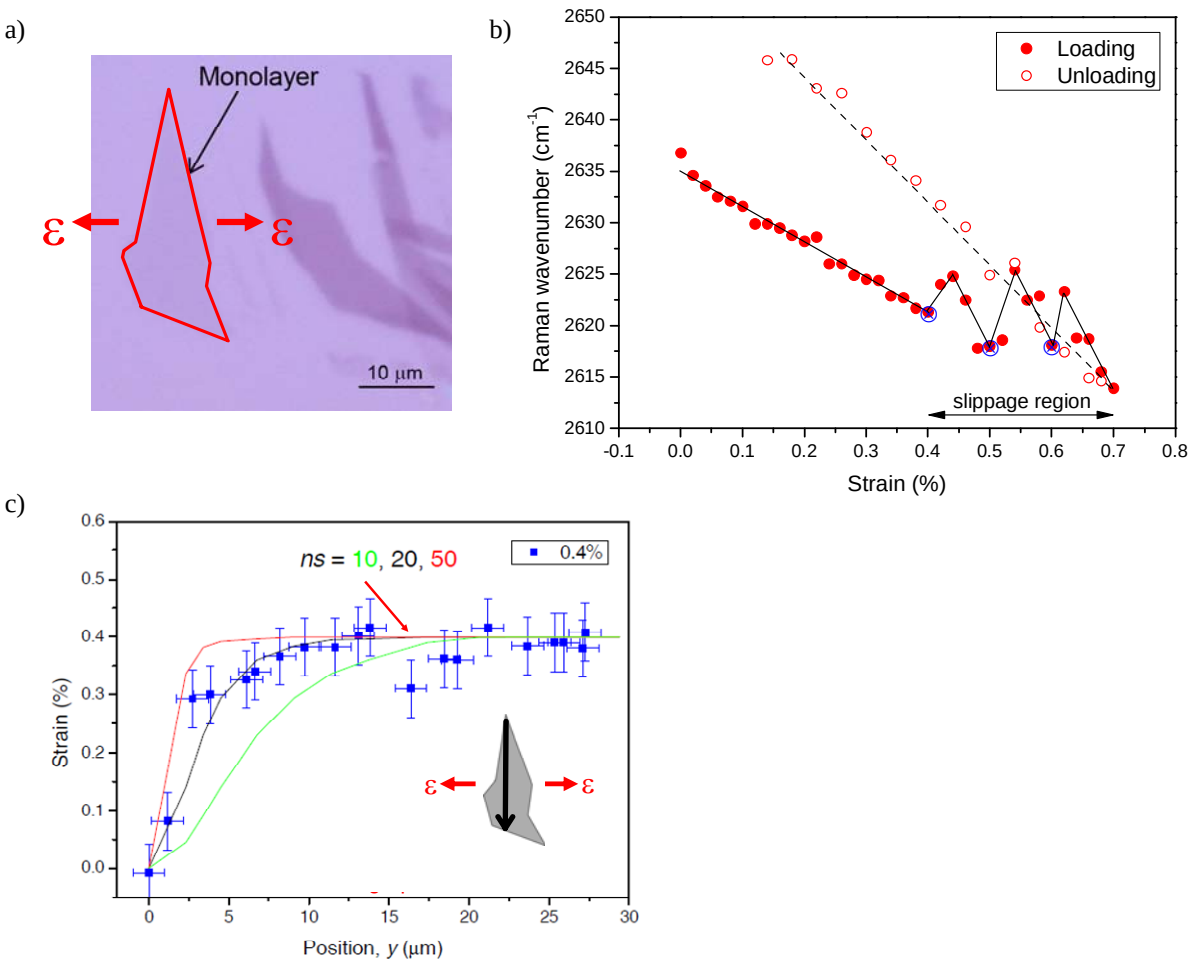

Figure 1a) An optical micrograph showing the flake studied for b) and c), with its outline and the direction the deformation was applied highlighted. b) The position of the G' band at the centre of the flake as function of global strain. The relaxation in the slippage region occurred during mapping of the strain in the flakes. c) The strain distribution down the centre of the flake, scanned perpendicular to the direction of the applied deformation. The curves show the fits of the shear lag model for different values of the factor of stress efficiency transfer, n, and aspect ratio, s.

References

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