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# Graphene resting on substrate: Closed form solutions for the perfect bonding and the delamination case

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ABSTRACT

We study closed form solutions for the perfect bonding and the delamination case for a monolayer graphene sheet resting on an elastic foundation. The theoretical framework we adopt is restricted to the materially and geometrically linear case. Graphene is modeled as a hexagonal 2-lattice, while the substrate is assumed to behave in an isotropic linearly elastic manner. Initially, we ignore out-of-surface motions and study the case of biaxial tension/compression and simple shear. We find the components of the shift vector by solving the equations ruling the shift vector. We then substitute this expression for the shift vector components to the momentum equation. This way we obtain conditions that the field of the internal strains, the strain constants and the material parameters should satisfy in order biaxial tension/compression and simple shear to be solutions for all equilibrium equations. For the particular case of axial strain and for the simple shear case we plot the mean stress components versus strain for three different substrates. Then, we take into account out-of-surface motions. We assume the out-of-surface displacement to be the product of a wave-like function and an unknown function, which we determine under certain conditions. These conditions are constraints that the field of the internal strains, the strain constant and the material characteristics of the substrate and graphene should satisfy in order the equilibrium equations to be satisfied. These cases pertain to the perfect bonding case. Distinguishing film's displacement from the bulk (substrate) displacement we study the case where delamination occur. We again use a semi-inverse method: we assume film's displacement to be the product of a wave-like function with an unknown function. The bulk's displacement is assumed to be different from the one of the film, in areas of delamination. We determine the unknown function present in the displacement of the film, by requiring the momentum equations to be satisfied.



## 1. Introduction

Graphene is a two-dimensional sheet that constitutes the building unit of all graphitic forms of matter, such as graphite, carbon nanotubes and carbon fibers. Graphene attract much attention to the mechanics community due to its very high strength of approximately 1 TPa (Lee et al., 2008). This together with its very small thickness, of approximately 0.335 nm, makes graphene an ideal potential candidate for strengthening composite structures. In a recent article we review the mechanical properties of graphene as probed by spectroscopic measurements and as calculated by ab initio, molecular simulation and continuum mechanical methods (Galiotis et al., in press).

On the other hand, graphene's very small thickness has some unpleasant consequences when trying to subject it to experiments: it is very difficult to grab graphene and apply some kind of loading. To remedy this situation workers embed graphene samples on a polymer substrate and load the system graphene/substrate. Then the technique of Raman spectroscopy can be applied to measure the mechanical properties of graphene by measuring the G and 2D peak of the Raman spectra. In Androulidakis et al. (2014) we embed a graphene flake on a substrate and apply a tensional loading to the system graphene/substrate using either the technique of cantilever beam or the four point bending technique.

The present work is motivated from the above described use of the substrate. It targets to mathematically model the graphene/-substrate system when subjected to simple loadings. In a sense, this is a continuation of our previous efforts to model free-standing graphene (Sfyris and Galiotis, in press; Sfyris et al., 2014a,b; Sfyris et al., 2015), by taking into account substrate's

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presence. In Sfyris and Galiotis (in press), Sfyris et al. (2014a,b) and Sfyris et al. (2015) we present the mathematical background for modeling graphene at the continuum level. In particular, adopting the framework of Steigmann and Ogden (1999) we utilize a surface free energy for graphene based on three arguments. The first argument is an in-surface stain measure describing changes taking place on the surface. The second argument is the curvature tensor which describe the out-of-surface motion and introduce bending into the model. The third argument is the shift vector. The motivation for assuming the shift vector as an argument stem from well established theories of crystalline materials (Parry, 1978; Ericksen, 1970; Ericksen, 1979; Fadda and Zanzotto, 2000; Fadda and Zanzotto, 2001; Pitteri, 1984; Pitteri, 1985; Pitteri and Zanzotto, 2003).

In this sense we stress that graphene is modeled as a hexagonal 2-lattice (Sfyris and Galiotis, in press; Sfyris et al., 2014a,b).The need for viewing graphene as a multilattice stem from the fact that graphene's lattice cannot be seen as a Bravais simple lattice. In standard terminology of applied crystallography (Fadda and Zanzotto, 2000; Fadda and Zanzotto, 2001; Pitteri, 1985), graphene's lattice belong to a special category of multilattices: it is a hexagonal 2-lattice. The unit cell for all possible plane 2-lattices is given in Fadda and Zanzotto (2000). The fact that graphene is at the discrete level a 2-lattice has some important consequences when scaling up to the continuum. The most important consequence is that the shift vector should be an independent argument at the continuum energy (Parry, 1978; Ericksen, 1970, 1979; Fadda and Zanzotto, 2000, 2001; Pitteri, 1984, 1985; Pitteri and Zanzotto, 2003). The shift vector is the vector connecting the two simple hexagonal lattices that constitute the hexagonal 2-lattice of graphene (see also the Figures in Sfyris and Galiotis, in press; Sfyris et al., 2014a,b; Sfyris et al., 2015). So, at the continuum level the energy should depend on the shift vector as well.

Lamdmark works on the continuum modeling of graphene stem from the fundamental work of Lee et al. (2008) who use a nanoidentation experiment in an atomic force microscope to measure the elastic properties and intrinsic strength of graphene. Using second order elasticity these authors evaluate Young's modulus, the second order elastic constant as well as graphene's breaking strength. Their analysis model graphene as an isotropic body in one dimension, due to symmetry in the loading. Generalization of their approach to two dimensions is done by Cadelano et al. (2009). These authors view graphene as an isotropic body and they utilize an energy cubic in strains (second order elasticity in words of Murnaghan (1951) and Rivlin (1963)). Utilizing tight-binding atomistic simulations they calculate Young's modulus, Poisson ratio as well as higher order constants for graphene. While interesting and novel their approach is, it lacks the treatment of bending effects. It also model graphene as an isotropic body; dependence on the zigzag and the armchair direction is not incorporated to the constitutive law through dependence on a structural tensor. Fifth order models for graphene are presented by Wei et al. (2009). These authors utilize an energy that depends on strains of the fifth order. Using density functional theory for simple loading histories they evaluate higher order constants for graphene. Their approach does not include bending effects neither anisotropy; graphene is modeled as an isotropic body.

Compared to these fundamental and interesting works, our line of work for modeling graphene as a 2-lattice (Sfyris and Galiotis, in press; Sfyris et al., 2014a,b; Sfyris et al., 2015) add novelty in three levels: a. we include bending effects into our analysis by dependence of the energy on the curvature tensor, b. symmetries of graphene are properly taken into account starting from the discrete picture and passing consistently to the continuum using the structural tensor in line with the classical theories of invariants of nonlinear elasticity, c. our analysis is devoid of the endless Taylor expansion of the energy the abovementioned works utilize: evaluating the invariants we find the exact number of material parameters for a most generic energy describing the material at hand. To all these we add that graphene is a monoatomic 2-lattice and not a simple lattice that almost all works in literature assume. Thus, at the continuum energy the shift vector should be taken into account in line with well established theories of multilattices (see e.g. Chapter 11 of Pitteri and Zanzotto (2003)).

In the literature there are many works related with thin film/-substrate interactions, from the theoretical point of view. We refer to the paper by Mishnaevsky and Gross (2005) for a concise review of this topic as well as to its numerous references. We draw particular attention to the fundamental paper by Huang (2005). There, the substrate behave viscoelastically while for the thin film the von-Karman assumptions are adopted. Using plane strain analysis and the standard Laplace transformation method for converting a viscoelastic problem to an elastic one, the author solve the viscoelastic problem of the substrate. The effect of the thin film is present on the boundary condition of the equations governing the bulk material.

Another interesting study is the approach of Cao and Hutchinson (2012) who adopt a neo-Hookean expression for the energy of the film as well as for the substrate in order to study the effect of the pre-stretch of the substrate. Fried and Todres (2005), study the effect of curvature and residual stress to the buckling of a half space with free surface near a contactor. They assume that the bulk and the boundary body are made of the same material and use the geometrical symmetry to reduce the problem to one dimension only. It is important to note that these authors introduce van der Waals effects in their analysis. The effect of the surface tension of a free surface on the bulk material is studied by Wang et al. (2010) who also assume that the free surface and the bulk body are made of the same material. The case of partial delamination of the thin film from the substrate is an undesirable phenomenon which appears frequently in the manufacturing process. Bedrossian and Kohn (2015) lay down a specific expression for the displacement function that describe partial delamination in the form of a blister.

The main novelty of the present contribution lies on the fact that we take into account the presence of the substrate on closed form solutions related with simple loadings. This is done for the case where graphene and substrate are perfectly bonded and also for the case where delamination take place. Additionally, we retain throughout the analysis terms related with residual strains for both the thin film and the substrate. We adopt the field equations as reported by Chhapadia et al. (2011). These are the momentum and the moment of momentum equations for the thin film (graphene) in the absence of body forces and inertia. The effect of the substrate enter through terms present in these equations. To these equations one should add the equation ruling the shift vector (Parry, 1978; Ericksen, 1970, 1979; Fadda and Zanzotto, 2000, 2001; Pitteri, 1984, 1985; Pitteri and Zanzotto, 2003) since graphene is a multilattices.

To bring our framework closer to more applied approaches and to give a rough order of magnitude for plotting purposes, we present the mean stress–strain diagrams for the axial tension test and the simple shear problem. For the substrate we make three different assumptions corresponding to three common materials used as substrate: Polyethylene terephthalate (PET), Polymethyl methacrylate (PMMA) and Polydimethylsiloxane (PDMS). In the mathematical model these are introduced through their Lame constants, $\lambda$, $\mu$. Being aware that these polymeric substrate's behave in a viscoelastic manner, we assume the viscous response to be negligible, since we have in mind experiments with slow rate such as those in Androulidakis et al. (2014).

For graphene we adopt the linear framework of Sfyris et al. (2014b), so the whole theory is confined to geometrical and