Ripples, Phonons and Bandgap in Strained Graphene

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***Abstract*— Using a novel interatomic force field, called MMP, we study the morphology of Graphene layers under a variety of strain conditions. We report that strain induced ripples possess the "right" kind of elastic deformation that is necessary in order to produce appreciable bandgap opening, which we calculate using Tight Binding, even for low enough strain that can be accessed through realistic means. At the same time the vibrational properties, calculated from analytic derivatives of the MMP force field and used within the dynamics matrix method, can be easily linked to strain obtained from Molecular Dynamics, opening the way for accurate modelling of Raman data. We also show that our models have allowed us to realize in practice novel devices based on our predictions.**

Keywords*— *Empirical interatomic potential; Tight Binding; Molecular Dynamics; Graphene.

I. INTRODUCTION

Two-dimensional (2D) layers like graphene are subject to long-wavelength fluctuations with strong height fluctuations often referred to as ripples or wrinkles. The specific shape, form and height of such ripples will in general depend on the external strain field present. We have therefore performed molecular dynamics (MD) of suspended graphene, by the use of a newly developed force field model (MMP)[1] that on top of having provided excellent results for Si and Ge [2], also proves to be extremely accurate for both C Diamond and Graphene [3]. The reason is that the MMP potential successfully reproduces the energy of the σ-bonds in both sp^3 and sp^2 configuration, despite being essentially a pair potential with the minimal inclusion of only some long range interactions. Here we present the results of MD simulations, compared to experimental electron microscopy analysis, in order to reveal how ordered and static ripples form spontaneously as a direct response to external pressure, although the strain present around the ripples can be very different form the externally present strain. As a surprising result, different regions of the strained graphene sheet, investigated by tight-binding, exhibit localized bandgap opening of the order of 0.2eV, suitable for far IR applications. Having reported controllable morphological changes as a function of strain, we can show how this provides the means to practically control and tune the electronic and transport properties of graphene for applications as optoelectronic and nanoelectromechanical devices.

II. THE MMP POTENTIAL

Atomistic empirical potential methods are still the only computationally feasible way to model semiconductor materials on nanoscale as they can drastically reduce the computational compared to ab initio methods.

Starting from the Abel-Tersoff model [4] [5], which is one of the most commonly used methods for atomistic modelling, the MMP [1] is a many body potential that simultaneously reproduces the elastic constants, dispersion curves and mode-Grüneisen parameters for the group IV semiconductors. The inclusion of radial and angular forces of the interacting atom pairs (short range) together with the partial inclusion of long range interactions are taken into account up to the second nearest neighbours, is enough to correctly influence the elastic and vibrational properties. In this way the original conundrum observed in the ATPs but also in others potential as the Stillinger-Weber [6] or the Tersoff-Brenner potential [7], i.e. the incompatibility between vibrational (phonon frequencies and their dispersions) and elastic properties (e.g. elastic constants) is finally resolved in the MMP potential.

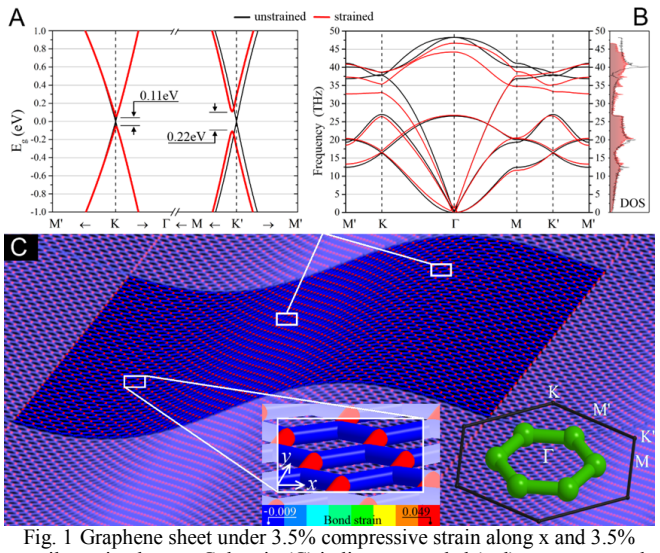

Fig. 1 Graphene sheet under 3.5% compressive strain along x and 3.5% tensile strain along y. Colors in (C) indicate extended (red) or compressed (blue) bonds. The bandstructure (A) exhibits a bandgap of 0.22eV at the K' point of the Brillouin zone.[3]

III. RESULTS AND DISCUSSION

Using MD we studied ripple formation of a graphene sheet under different strain conditions. These include both tensile and compressive pressures. The MD simulations, comprising around 4000 atoms, use as a starting point an atomically flat single graphene sheet. Pressure is then simulated by rigid displacement of the atoms on the xy plane. The initial structure is therefore a strained graphene sheet, energy minimization is then used to determine the relaxed atomic

structure. Here we show the two cases of compressive strain in the x direction of 3.5% together with tensile strain in the y direction of 3.5% (Fig. 1), and also, compressive strain of 3.5% in both the x and y directions (Fig. 2).

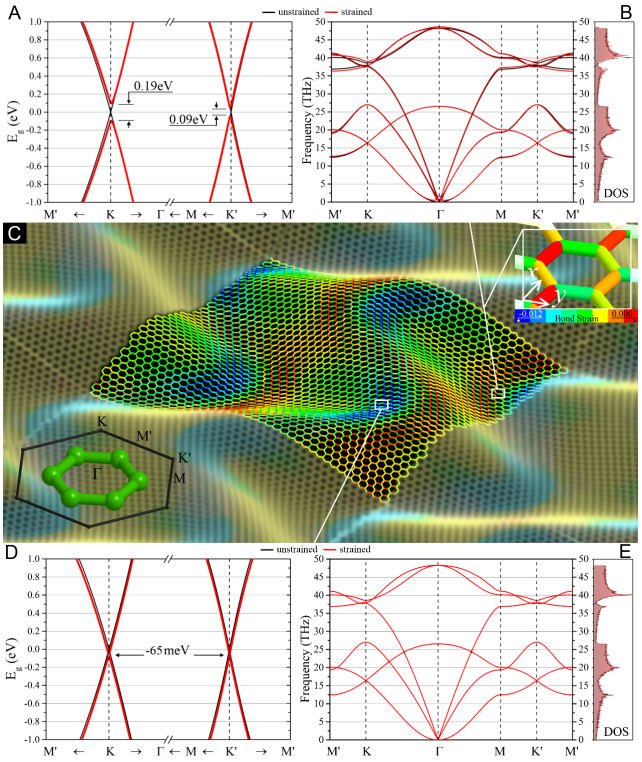

Fig. 2. Graphene sheet under 3.5% hydrostatic compressive strain. Colors in (C) indicate extended (red) or compressed (blue) bonds. The bandstructure (A) at the Dirac points as calculated by tight binding predicts a bandgap of 0.19eV at the K point of the Brillouin zone in the regions of tensile (red) strain. Regions of compressive (blue) or low (to zero) strain (green) in (C) do not exhibit a bandgap (D) or changes in the phonon spectrum (E) compared to the unstrained case. However for compressive (blue) strain (D) the Dirac point lies below the Fermi energy by 65meV thus exhibiting spontaneous n-doping behavior.[3]

In Fig. 1 and Fig. 2 we also show the tight binding calculated bandstructure in the proximity of the Dirac point for particular areas (unit cells) of the graphene sheet. The red lines are compared to the unstrained case (black lines). Non hydrostatic strain (e.g. shear or internal displacement) introduces non equivalencies in the high symmetry points, so that the K and M points are no longer degenerate and both the band structure and phonon dispersions need to be described with the addition of the K' and M' points. For both types of ripples the bandgap opens, by 0.22eV (0.11eV) near the K' (K) point for the wave front ripple and by 0.19eV (0.09eV) at the K (K') point for the sinusoidal ripple. In this case this only happens in the regions with predominantly tensile strain. In the compressive strain regions the most notable feature is that compared to the unstrained case the Dirac point is found below the Fermi energy by 65meV, thus exhibiting strain activated spontaneous n-doping characteristics.

The results of the simulations are compared with the results of both μRaman, transmission electron microscopy and atomic force microscopy (Fig. 3). The experimental results show remarkable features which closely match the structural modelling predictions, therefore suggesting that the predicted electronic properties can also be realized in real structures.

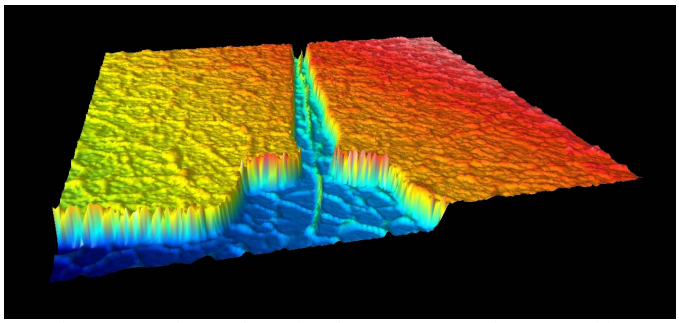

Fig. 3. AFM of a Graphene sheet between two metal contacts with a separation of 1μm. The Graphene is subjected to compressive strain resulting in the formation of a stable nm-sized ripple in the middle of the gap.

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