Tension buckling of graphene: A new phenotype

Y. Xiang [a], Hui-Shen Shen [b],*

[a] School of Computing, Engineering and Mathematics, University of Western Sydney, Locked Bag 1797, Penrith, NSW 2751, Australia
[b] School of Aeronautics and Astronautics, Shanghai Jiao Tong University, Shanghai 200240, People's Republic of China

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ABSTRACT

We report here a new phenotype of buckling of a monolayer free hanging graphene subjected to uniform distributed tension force on all atoms by molecular dynamics simulations. We find that buckling occurs when tension force is applied and is sufficiently large for both armchair and zigzag monolayer graphene sheets. The maximum amplitude oscillates around 3.60 nm for the free hanging zigzag graphene, but is decreased for the free hanging armchair graphene; whereas the associated applied tension force is decreased for both armchair and zigzag graphene sheets when the temperature is increased.

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1. Introduction

The first isolated graphene flakes were successfully produced by Geim and Novoselov in 2003 [1–3] which earned them the prestigious 2010 Nobel Prize in Physics. Graphene is an one-atom-thick and two-dimensional layer consisting of sp^2-bonded carbon atoms and is one of the strongest and stiffest known materials. Graphene not only possesses extraordinary mechanical properties, but also has outstanding electronic, chemical and optical characteristics and has vast potential applications in various scientific and engineering fields. Due to the sp^2-bonding amongst the carbon atoms, ideally these atoms in a graphene form perfect flat hexagonal lattice in a two-dimensional platform. However, due to the influence of heat fluctuations, graphene cannot maintain its 2D flat form and elastic corrugations (ripples) are formed to maintain the stability of the graphene as first reported by Meyer et al. [4] for a free-hanging graphene. Carlsson [5] reviewed several studies on the behavior of two-dimensional crystals, including graphene, in finite temperature environment and concluded that ripples or thermal buckling are an intrinsic feature of graphene [6,7]. Fasolino et al. [8] studied an infinitely large graphene sheet in an environmental temperature of 300, 1000, 2000 and 3500 K by the Monte Carlo simulations and observed that ripples were formed to maintain the equilibrium of the graphene due to thermal fluctuations. Abedpour et al. [9] also observed ripples in graphene and they suggested that it is due to the negative thermal expansion

coefficient (TEC) of graphene. This may not be able to explain ripples in graphene at very high temperatures as the negative thermal expansion coefficient of graphene was suggested to be in the range of 0–450 K [10–12]. Other researchers studied the buckling of graphene by inducing in-plane compressive force at the edge of the graphene and most of the studies are based on continuum mechanics theories [13–18]. Baimova et al. [19] investigated the compressive strain induced ripples in clamped graphene nanoribbons. From the theory of elastic stability, there are in general two ways to induce buckling in a thin film: (1) stretching the thin film in the axial direction, buckling occurs perpendicular to the axial direction, or (2) compressing or displacing the thin film in the axial direction, wrinkling (i.e. local short wavelength buckling) occurs along the axial direction. Shen et al. [20] conducted a study on a free standing graphene subjected to tensile forces on two opposite edges using the molecular dynamics (MD) simulations. They observed that tension buckling does occur in both armchair and zigzag graphene sheets when the tension force is sufficiently large. Another study on tension buckling of graphene was performed by Wang et al. [21] in which the two opposite edges of the graphene are clamped and a set of local tension forces are applied on the edges of the graphene. However, the first observed buckling is for free-hanging graphene [4] and we still need to understand why buckling occurs for a monolayer free-hanging graphene. The present paper extends the previous work [20] to the case of the tension buckling of a monolayer free hanging graphene. The tension force is assumed to be uniformly distributed and is applied on each atom in the graphene instead of the tension forces on the two opposite edges, as reported in Shen et al. [20].

* Corresponding author.
  E-mail address: hsshen@mail.sjtu.edu.cn (H.-S. Shen).

2. Modeling and simulations

Two types of monolayer graphene sheets, i.e. armchair and zigzag graphene sheets, are considered. The definitions of armchair and zigzag are the same as those used in carbon nanotubes [10,22]. The thermal environmental conditions are assumed to be at $T=300$, 400, 500, 600 and 700 K. The large-scale atomic/molecular massively parallel simulator (LAMMPS from http://lammps.sandia.gov/index.html) [23] is used in our simulation with the adaptive intermolecular reactive empirical bond order potential (AIREBO) [24] being adopted for the system of carbon atoms in the graphene. The AIREBO potential consists of three potential components and can be expressed as [24]

$$E=\frac{1}{2}\sum_i\sum_{j\neq i}\left[E_{ij}^{REBO}+E_{ij}^{LJ}+\sum_{k\neq i,j}\sum_{l\neq i,j,k}E_{kijl}^{TORS}\right] \quad (1)$$

where E_{ij}^{REBO} is the potential for pairwise covalent bonding interactions of atoms based on the 2nd generation reactive empirical bond order potentials of Brenner, E_{ij}^{LJ} is the potential for the pairwise sum of Lennard–Jones interactions and E_{kijl}^{TORS} is the potential for the torsion interactions depending on the dihedral angles of the system. The Nose–Hoover thermostat is employed in the simulation to maintain the temperature of the system [25].

The considered armchair graphene is of the length $L_x=15.197$ nm and the width $L_y=14.637$ nm, while the length and width of the zigzag graphene are reversed as $L_x=14.637$ nm and $L_y=15.197$ nm, respectively. There are totally 8640 carbon atoms in either the armchair or the zigzag graphene. To achieve the free hanging effect, only the top row of atoms in the graphene is constrained in the y direction while these atoms are free to move in the x and z directions. This constraint condition is enforced through the LAMMPS command "fix 10 upper drag NULL 146.37 NULL 12 0.01" for the armchair graphene, where 10 and upper are the ID and the top row atom group-ID respectively, drag is the style of the fix command, 146.37 is the y coordinate of the top row atoms in Å, and NULL, 12 and 0.01 are the parameters required by this fix command.

We take a zigzag graphene under the environmental temperature 400 K as an example to illustrate the simulation process. We first test that the ensemble of velocities of the atoms in the graphene is generated at 400 K using the Gaussian distribution and then the top row of atoms in the graphene is constrained as described previously. When performing the energy minimization of the system with stopping tolerance for energy being set as 10^{-8}, we observe that a few bonds between the top row and the second top row atoms are damaged due to high initial velocities of the atoms during the minimization process. In order to overcome this problem, we reduce the initial thermal kinematic energy of the atoms by setting the ensemble of velocities of the atoms at 100 K. The energy minimization of the system is performed and the temperature of the system is then ramped up to 400 K in 16,000 time steps with each time step being set as 0.1 fs. No bondage breakage is observed during this process and further MD simulations are carried out as follows. A tensile force 4.89×10^{-4} nN (along the negative y direction) is then applied on each atom in the graphene. The positions and velocities of the atoms in the system are updated in 2000 runs with a time step for each run being set as 1 fs while the environmental temperature of the system is kept around 400 K through the Nose–Hoover thermostat. The loading process is repeated until a large number of bonding breakages occur and the graphene can no longer sustain the tensile force.

3. Results and discussion

First, we present a case for a free hanging zigzag graphene under 400 K environmental temperature to show the different
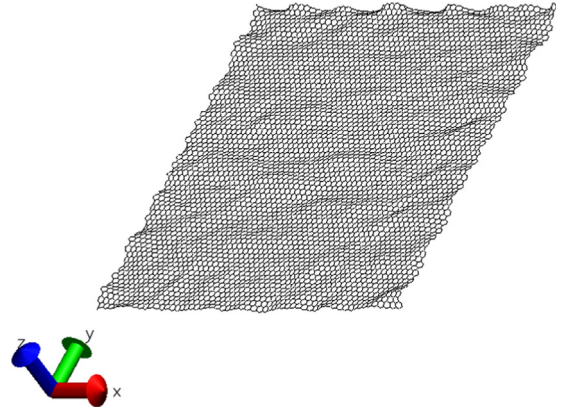

Fig. 1. (Color online) The state of a free hanging zigzag graphene at $T=400$ K before tensile force is applied.
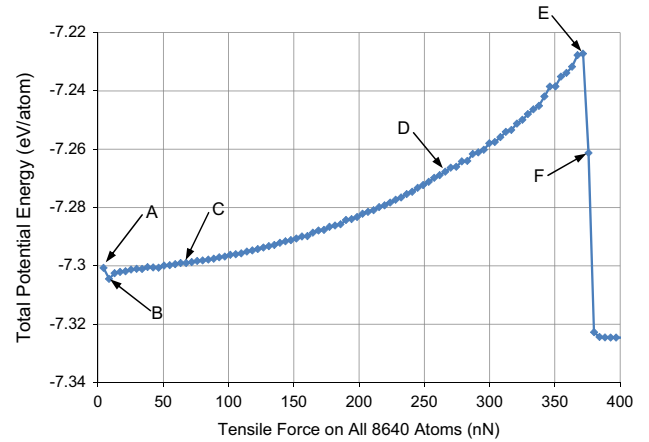

Fig. 2. (Color online) Total potential energy versus tensile force for a free hanging zigzag graphene at $T=400$ K.

stages of the MD simulation and associated results. Fig. 1 presents the state of the graphene at the end of the temperature ramp from 100 to 400 K and before any tensile force is applied on the atoms. We observe that the graphene achieves stability through the formation of finite size ripples due to thermal fluctuations. We do not regard this state of the graphene as buckling as the ripples and the corresponding waves are not as regular as the ones in a buckled thin sheet at this stage. In this study, we define that the tension buckling state of a free hanging graphene is that regular waves along the x direction appear at the lower edge of the graphene while the two side edges are relatively flat under the tensile force.

Fig. 2 shows the variation of the total potential energy of the graphene system against the total tensile force with the loading process as described in Section 2. Point A and point B on the curve in Fig. 2 are corresponding to the two initial loading steps with total tensile force being 4.22 nN and 8.45 nN, respectively. We observe that the total potential energy of the graphene decreases slightly when the tensile force is increased from 4.22 to 8.45 nN. The total energy of the graphene increases after point B and reaches its peak at point E which is corresponding to the maximum tensile force that the graphene can sustain. The total energy of the graphene has a sudden drop after point E which indicates that the graphene is damaged and can no longer sustain the tensile force.

The states of the graphene corresponding to the two initial loading steps are depicted in Fig. 3. It is seen that the application of a small initial tensile force (4.22 nN) has created larger ripples on the graphene when compared with the state of the graphene