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The effects of free edge interaction-induced knotting on the buckling of monolayer graphene

Hao-Yu Zhang[a], Jin-Wu Jiang[a,*], Tienchong Chang[a], Xingming Guo[a], Harold S. Park[b]

[a] Shanghai Institute of Applied Mathematics and Mechanics, Shanghai Key Laboratory of Mechanics in Energy Engineering, Shanghai University, Shanghai 200072, People's Republic of China
[b] Department of Mechanical Engineering, Boston University, Boston, MA 02215, USA

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ABSTRACT

Edge effects play an important role for many properties of graphene. While most works have focused on the effects from isolated free edges, we present a novel knotting phenomenon induced by the interactions between a pair of free edges in graphene, and investigate its effect on the buckling of monolayer graphene. Upon compression, the buckling of graphene starts gradually in the form of two buckling waves from the warped edges. The collision of these two buckling waves results in the creation of a knot structure in graphene. The knot structure enables the buckled graphene to exhibit two unique post-buckling characteristics. First, it induces a five-fold increase in graphene's mechanical stiffness during the buckling process. Second, the knotted structure enables graphene to exhibit a mechanically stable post-buckling regime over a large (3%) compressive strain regime, which is significantly larger than the critical buckling strain of about 0.5%. The combination of these two effects enables graphene to exhibit an unexpected post-buckling stability that has previously not been reported. We predict that numerical simulations or experiments should observe two distinct stress strain relations for the buckling of identical graphene samples, due to the characteristic randomness in the formation process of the knot structure.

1. Introduction

Graphene is a quasi two-dimensional (2D) honeycomb lattice structure that exhibits extremely high in-plane stiffness (Lee et al., 2008) but very small bending stiffness (Ou-Yang. et al., 1997; Tu and Ou-Yang, 2002; Arroyo and Belytschko, 2004; Lu et al., 2009). The quasi 2D nature of graphene is the origin for many of the interesting phenomena involving graphene, including edge effects and buckling instability, which are of relevance to the present work.

For the buckling instability, Euler buckling theory (Timoshenko and Woinowsky-Krieger, 1987) states that the critical compressive strain, above which graphene is buckled, is inversely proportional to the in-plane stiffness C_{11} and is proportional to the bending stiffness D; i.e., $\epsilon_c \propto D/C_{11}$. According to Euler buckling theory, the critical strain for graphene is very small. Consequently, the buckling process can be induced by very weak external disturbances such as thermal expansion (Bao et al., 2009). As a result of the buckling phenomenon, graphene is bent or folded with a finite curvature, which can be used to manipulate many of its physical properties (Cong and Yu, 2014). As a result, the buckling of graphene has attracted intensive research interest in the past few years (Lu and Huang, 2009; Patrick, 2010; Sakhaee-Pour, 2009; Pradhan and Murmu, 2009; Pradhan, 2009; Frank et al., 2010; Farajpour et al., 2011; Tozzini and Pellegrini, 2011; Rouhi and Ansari, 2012; Giannopoulos, 2012; Neek-Amal and Peeters, 2012; Shen et al., 2013). Besides graphene, a group of other quasi 2D materials, eg. MoS_2 or black phosphorus, also have small critical buckling strains because the bending stiffnesses for these atomically thin materials are also fairly small (Jiang, 2014, 2015; Jiang et al., 2013).

As another result of graphene's 2D nature, edge effects play an important role on its physical properties. Based on the Brenner interatomic potential (Brenner et al., 2002) and the finite element method, it was demonstrated that graphene's free edges can become warped due to the compressive edge stress (Shenoy et al., 2008). The warping amplitude decays exponentially from the edge into the center; i.e., the height (z) of the warped configuration is $z \propto e^{y/l_c}$ with l_c as the critical penetration depth. The critical penetration depth can be viewed as the size of the warped edge region. For narrow graphene nanoribbons, the size of the edge region can be comparable to or larger than the central region.

If the size of the edge region in graphene nanoribbons is sufficiently large, the free edges dominate most of graphene's physical properties. Edge reconstructions have been observed

* Corresponding author.
E-mail address: jiangjinwu@shu.edu.cn (J.-W. Jiang).

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experimentally (Gass et al., 2008), which can be attributed to the thermal energy localized by the edge vibrations (Jia et al., 2009; Engelund et al., 2010). Edge vibrations were also found to be responsible for the larger energy dissipation in graphene nanomechanical resonators (Kim and Park, 2009; Jiang and Wang, 2012). It was found that edge effects are the dominant factor for the friction between neighboring nanotubes in multi-wall carbon nanotubes (Guo et al., 2011), and a piece of graphene can be driven from a softer regime to the stiffer regime due to the edge effect (Chang et al., 2015). Edge effect have recently been shown to induce negative Poisson's ratio in graphene nanoribbons with widths smaller than about 10 nm (Jiang and Park, 2016). While we have listed just a few examples here, free edges also have a strong effect on other physical properties in graphene (for a review, see eg. Ref. Castro Neto et al. (2009)).

Although edge effects on the mechanical properties in graphene have been extensively studied, the edge effect on buckling has not been examined to-date. Furthermore, free edges almost always are present in pairs. However, in the aforementioned works, each free edge makes an independent contribution to those mechanical properties in graphene. If the width of the graphene is comparable with twice the critical penetration depth l_c, there should be a strong correlation and interactions between the pair of free edges. The effect from a pair of correlated edges on the mechanical properties of graphene has not been studied yet. We thus investigate the effect from a pair of correlated edges on the buckling phenomenon in graphene.

In this paper, we investigate the buckling process for graphene with a pair of free edges. Different from the usual abrupt buckling mode, we find that graphene is gradually buckled starting from the free edges if the two edges are warped in opposite directions. The gradual buckling is due to the formation of a knot structure that results from the collision of the buckling waves from the two edges. There are four major features brought by the knotting effect. (1) Graphene with knotted structure has a much higher mechanical stiffness than graphene without knotting during the buckling process. (2) It is more difficult to buckle narrower graphene nanoribbons with the knotted structure as the knot is stronger in narrower graphene. (3) As a result of the randomness in the knotting phenomenon, we predict that numerical simulations or experiments should observe two different buckling processes even for identical graphene samples with free edges. (4) The knot is formed by the collision of buckling waves from the two free edges, and the knot structure will be unknotted if the compressive strain is larger than a critical unknotting strain value. After unknotting, all graphene with different boundary conditions have the same final buckled structure.

2. Simulation details

The interaction between carbon atoms in graphene is described by the second generation Brenner potential (Brenner et al., 2002). For stretching or compression, the edges of the graphene in the strain direction, i.e. the +x and −x edges in Fig. 1(a), have prescribed motion in the strain direction only, while free boundary conditions (FBC) are used in the out-of-plane direction. Before tension or compression, the system is thermalized to a targeted pressure and temperature within the NPT (i.e. the particles number N, the pressure P and the temperature T of the system are constant) ensemble for 200 ps. The Nosé–Hoover (Nose, 1984; Hoover, 1985) thermostat is used for maintaining constant temperature and pressure. After thermalization, graphene is stretched or compressed in the x-direction in Fig. 1(a) by uniformly deforming the simulation box in this direction, while the structure is allowed to be fully relaxed in lateral directions during mechanical loading. The standard Newton equations of motion are integrated in time using the velocity Verlet algorithm with a time step of 1 fs. Molecular dynamics (MD) simulations are performed using the publicly available simulation code LAMMPS (Plimpton, 1995; Lammps, 2012). The OVITO package was used for visualization (Stukowski, 2010).

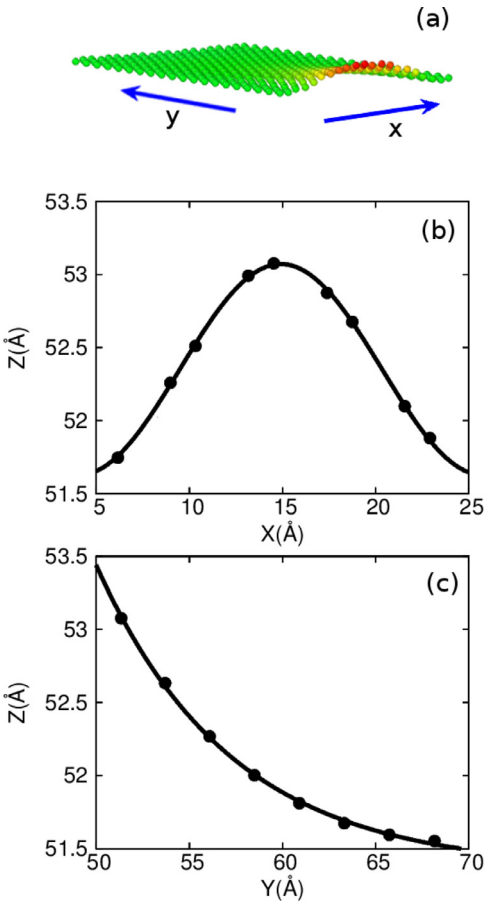

Fig. 1. (Color online) Warped configuration at 1 K of a free edge in graphene of dimension 30 × 80 Å. Half of the system is shown in the figure, while the other half (with another warped edge) is not shown. (a) Perspective view of the warped edge. The warped shape is described by the function $z(x, y) = z_0 + A\sin(\pi x/L)e^{-y/l_c}$. (b) z-position for atoms at $y = y_{\min}$. (c) z-position for atoms at the middle plane $x = 15$ Å. The color is with respective to the z-position of each atom. Graphene is compressed or stretched in the x-direction.

3. An isolated edge

3.1. Warped configuration

It has been demonstrated that free edges are warped due to the compressive edge stress in graphene (Shenoy et al., 2008). A typical warped edge configuration is illustrated in Fig. 1(a). The dimension of the graphene is 30 × 80 Å. The two ends in the x-direction are fixed, while FBC is applied in the y-direction. Only half of the system is shown, while the other warped edge is not displayed. The structure is relaxed at 1.0 K. The warping amplitude decays exponentially from the free edge into the center. Fig. 1(b) and (c) show that the height (z) of each atom can be well described by the function $z(x, y) = z_0 + A\sin(\pi x/L)e^{-y/l_c}$, where $L = 30$ Å is the length of graphene along the x-direction. Fitting parameter A is the warping amplitude, and $l_c = 7.3$ Å is the critical penetration depth of the warping edge.

An isolated free edge can be warped either in the +z or −z direction, whose structures are denoted by $\eta = \pm 1$ in Fig. 2, and whose corresponding configurations are displayed as the two

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