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Understanding and strain-engineering wrinkle networks in supported graphene through simulations

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ABSTRACT

Wrinkle networks are ubiquitous buckle-induced delaminations in supported graphene, which locally modify the electronic structure and degrade device performance. Although the strong property–deformation coupling of graphene can be potentially harnessed by strain engineering, it has not been possible to precisely control the geometry of wrinkle networks. Through numerical simulations based on an atomistically informed continuum theory, we understand how strain anisotropy, adhesion and friction govern spontaneous wrinkling. We then propose a strategy to control the location of wrinkles through patterns of weaker adhesion. This strategy is deceptively simple, and can in fact fail in several ways, particularly under biaxial compression. However, within bounds set by the physics of wrinkling, it is possible to robustly create by strain a variety of wrinkle network geometries and junction configurations. Graphene is nearly unstrained in the planar regions bounded by wrinkles, highly curved along wrinkles, and highly stretched and curved at junctions, which can either locally attenuate or amplify the applied strain depending on their configuration. These mechanically self-assembled networks are stable under the pressure produced by an enclosed fluid and form continuous channels, opening the door to nano-fluidic applications.

1. Introduction

Rather than a flatland, supported graphene describes a landscape shaped by out-of-plane features with different physical origins. Defects such as dislocations or grain boundaries can relax through out-of-plane deformations (Liu and Yakobson, 2010; Yakobson and Ding, 2011; Zhang et al., 2014), which have been observed experimentally (Duong et al., 2012; Warner et al., 2013). Gas trapped between the graphene and the substrate, either unintentionally (Stolyarova et al., 2009; Georgiou et al., 2011) or in a controlled manner (Bunch et al., 2008; Koenig et al., 2011; Zabel et al., 2012; Pan et al., 2012; Kitt et al., 2013), can elastically deform graphene, producing blisters of various shapes and sizes. Lateral strain produced upon cooling of graphene grown by chemical vapor deposition (CVD) on solid metallic substrates invariably results in linear and localized wrinkles (Li et al., 2009; Robertson et al., 2011; Obraztsov et al., 2007; Zhu et al., 2012; Liu et al., 2012), which persist after transfer to other substrates. Similar wrinkles have also been reported in exfoliated flakes (Xu et al., 2009). The large elastic strain caused by wrinkles or blisters disrupts the electronic structure of pristine graphene (Xu et al., 2009; Levy et al., 2010; Zabel et al., 2012), and therefore interferes with electronic transport (Zhu et al., 2012), plasmon propagation (Garcia-Pomar et al., 2013), and locally modifies chemical reactivity (Srivastava et al., 1999; Ruoff, 2012). For this reason, blisters and

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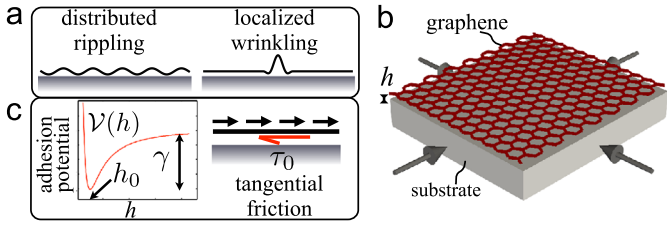

Fig. 1. (a) Upon compression, confined thin films develop a variety of out-of-plane deformations, classified here as *rippling*, distributed wave-like disturbances, or *wrinkling*, localized out-of-plane features surrounded by planar regions. (b) Model setup for a graphene flake adhered to a substrate and laterally compressed. h is the graphene–substrate separation. (c) Graphene interacts with the substrate through an adhesion potential $\mathcal{V}(h)$, Aitken and Huang (2010) characterized by the adhesion energy γ and the equilibrium separation h_0, and through tangential forces opposing sliding, modeled as dry friction with interfacial shear strength τ_0 (Zhang and Arroyo, 2013).

wrinkles are generally perceived as defects in graphene-based electronics (Zhu et al., 2012; Jiang et al., 2013) and optics (Garcia-Pomar et al., 2013). It has also been suggested that the strong coupling between localized deformation and electronic structure could be exploited by strain engineering (Levy et al., 2010; Castellanos-Gomez et al., 2013; Ruoff, 2012; Pereira et al., 2010; Zabel et al., 2012).

Here, we focus on wrinkles in supported graphene, generally understood as buckle-induced delaminations (Vella et al., 2009) caused by lateral compression. By intentionally straining the substrate uniaxially, it has been shown that lateral strain is transmitted to graphene by frictional forces, leading to localized wrinkling of monolayer graphene perpendicular to the strain direction (Jiang et al., 2013). Theoretical calculations suggest that, upon uniaxial compression, the localized wrinkles in graphene are preceded by distributed ripples of very small amplitude (Zhang and Arroyo, 2013), see Fig. 1(a). Most wrinkles observed in CVD graphene form two-dimensional networks (Li et al., 2009; Robertson et al., 2011; Obraztsov et al., 2007; Zhu et al., 2012; Liu et al., 2012), indicative of biaxial compressive strain and suggestive of stress focusing (Cerda et al., 1999; Witten, 2007; Pereira et al., 2010; Aoyanagi et al., 2010). Beyond isolated wrinkles, massive crumpling and delamination have been reported in supported multilayer graphene under very large biaxial compression (Zang et al., 2013).

A number of experimental strategies have attempted to control wrinkle networks in graphene. It has been shown that the transfer process can increase, decrease, or even eliminate wrinkling (Liu et al., 2011; Calado et al., 2012), and that wrinkles preferentially form at topographical features of the substrate (Kim, K. et al., 2011; Pan et al., 2011; Liu et al., 2012). However, it has not been possible to precisely and reversibly control the location of wrinkles, partly due to an insufficient theoretical understanding of wrinkling under biaxial compression (Zhu et al., 2012). Here, through detailed simulations (Section 2), we examine the spontaneous formation of wrinkle networks under biaxial strain (Section 3), identify the key governing mechanisms including the role of junctions, and exploit this understanding to propose a strategy to precisely control the network geometry through patterns of weaker graphene-substrate adhesion (Section 4). Since wrinkles exhibit anisotropic electronic transport, are obstacles to plasmons, and form nano-fluidic channels, our results may enable constructive use of strain-engineered wrinkle networks. We delineate the physical limits of this strategy, and characterize the behavior of different networks for strain-engineering and nano-fluidic applications.

2. Model and simulation method

We consider a mesoscopic (500×500 nm^2) graphene sample supported on a substrate undergoing lateral deformation under periodic boundary conditions, see Fig. 1(b). The domain is large enough to capture the typical wrinkle separation (several 100 nm) but small enough to be computationally tractable, since the geometric features of an individual wrinkle (~ 1 nm) (Zhu et al., 2012; Liu et al., 2012) need to be resolved.

2.1. Model for graphene

We model graphene with an atomistic-based continuum theory, built from Brenner's force field and discretized with subdivision finite elements (Arroyo and Belytschko, 2002, 2004b). This method has been shown to accurately simulate carbon nanotubes (Arias and Arroyo, 2008) and graphene (Zhang and Arroyo, 2013; Ghosh and Arroyo, 2013) undergoing large deformations and buckling instabilities at a fraction of the computational cost of an atomistic model. In this continuum model, we describe the configuration of graphene by a surface parametrization $\boldsymbol{x}(u,v) = (x(u,v), y(u,v), z(u,v))$, where $z(u,v)$ is the coordinate perpendicular to the substrate. The elasticity of this continuum description of graphene is modeled through a hyper-elastic potential of the form $W(\boldsymbol{C}, \mathcal{K})$ that depends on the in-plane strain (the Cauchy–Green deformation tensor) of the surface $\boldsymbol{C}$ and its curvature (the second fundamental form) $\mathcal{K}$. The energy density W is systematically derived from the atomistic model describing the bonding energy and forces using a kinematic rule that links continuum and lattice deformations, the exponential Cauchy–Born rule, and averaging the atomistic energy in one unit cell of the lattice. The total