

# Nanomechanical Architectures—Mechanics-Driven Fabrication Based on Crystalline Membranes

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## Abstract

Bending of thin sheets or ribbons is a ubiquitous phenomenon that impacts our daily lives, from the household thermostat to sensors in airbags. At nanometer-scale thicknesses, the mechanics responsible for bending and other distortions in sheets can be employed to create a nanofabrication approach leading to novel nanostructures. The process and resulting structures have been aptly referred to as “nanomechanical architecture.” In this article, we review recent progress in atomistic simulations that not only have helped to reveal the physical mechanisms underlying this nanofabrication approach, but also have made predictions of new nanostructures that can be created. The simulations demonstrate the importance of the atomic structure of the crystalline membrane and of the intrinsic surface stress in governing membrane bending behavior at the nanoscale and making the behavior fundamentally distinct from that at the macroscale. Molecular dynamics simulations of the bending of patterned graphene (a single-atomic layer film) suggest a new method for synthesizing carbon nanotubes with unprecedented control over their size and chirality.

## Introduction

Mechanical properties of materials encompass at the most fundamental limit parameters such as elastic constants and modulus, and at the empirical end quantities such as strength, hardness, and toughness. Because of the great importance of mechanical properties in the engineering of materials, an immense amount is known for bulk materials and even thin films and materials at the mesoscale, for example, in microelectromechanical systems (MEMS) devices. In nanostructures, the manifestation of a material’s mechanical properties may, however, differ significantly from that in its macroscopic counterparts.<sup>1–3</sup> Two limits can be considered. In the limit that the

nanostructure is so small that its properties are dominated by surfaces, the fundamental elastic constants themselves may change, producing quite novel mechanical effects. But even if the bulk elastic constants still dominate, many distinctive nanoarchitectures can be fabricated by (1) taking advantage of thinness and other nanodimensions, (2) judiciously combining materials, (3) manipulating the strain, and (4) taking advantage of the anisotropies of the elastic properties.<sup>4,5</sup> These nanoarchitectures, ranging from strained flat films to tubes, coils, rings, and “rug wrinkles,” in turn, enable not only significant new nanotechnologies but also serve to elucidate

mechanics at these dimensions. Our focus in this article is on simulations of nanofabrication via nanomechanics, within the overall theme of “atomistic simulations of nanomechanics.”

At the foundation of much of nanoarchitecture formation is strain, in particular, differential strain in two materials in contact or that emanate from two sides of a very thin sheet. A common device, the bimetallic-strip switch in a thermostat, illustrates differential strain: the strip bends because of different thermal stress in the two metals, which opens or closes the switch. Similarly, the combination of two thin materials in an epitaxial relationship produces a strained bilayer that bends because of lattice misfit strain in the two layers (even without considering temperature), an effect that is not observable if one of the materials is thick. In fact, when the thicknesses of the two layers are reduced to the nanometer scale, the bending magnitude of the bilayer can be so large that it can roll into tubular shapes with a characteristic radius of curvature that is also nanometer scale. The use of directed lattice misfit strain-induced distortion in bilayers leads to the novel nanomechanical architectures fabrication discussed in this article.<sup>1,4,5</sup>

The approach has several important technological advantages. It allows for the creation of nano- and microstructures with inexact dimensionality, ranging from micro- and nanorings (Figure 1a)<sup>5</sup> to coils (Figures 1b<sup>6</sup> and 1d<sup>5</sup>), tubes (Figures 1c,<sup>7</sup> 1e,<sup>8</sup> and 1f<sup>9</sup>), curved sheets,<sup>3</sup> or ribbons with odd shapes.<sup>10</sup> These different nanoarchitectures can be made based on *a priori* theoretical designs,<sup>5</sup> while their size and shape can be tuned over a wide range by choosing different combinations of materials, varying film dimensions, and applying external forces. The approach is extremely versatile, applicable to most materials combinations, including semiconductors (Figures 1a,<sup>5</sup> 1d,<sup>5</sup> and 1f<sup>9</sup>), metals (Figure 1b),<sup>6</sup> insulators (Figure 1e),<sup>8</sup> and polymers (Figure 1c).<sup>7</sup> It is also completely compatible with processing methods developed for Si, III–V, or MEMS technologies, and is thus suitable for parallel mass production of identical or different nanostructures (Figure 1f).<sup>9</sup>

The basic design principle and qualitative behavior of nanoarchitectures can be understood by classical mechanics and continuum theory.<sup>5</sup> Nevertheless, atomistic simulations have made significant contributions in revealing the prominent role played by the atomic nature of the membrane structure and by the intrinsic surface stress in governing the bending of nanomembranes. Multiscale modeling

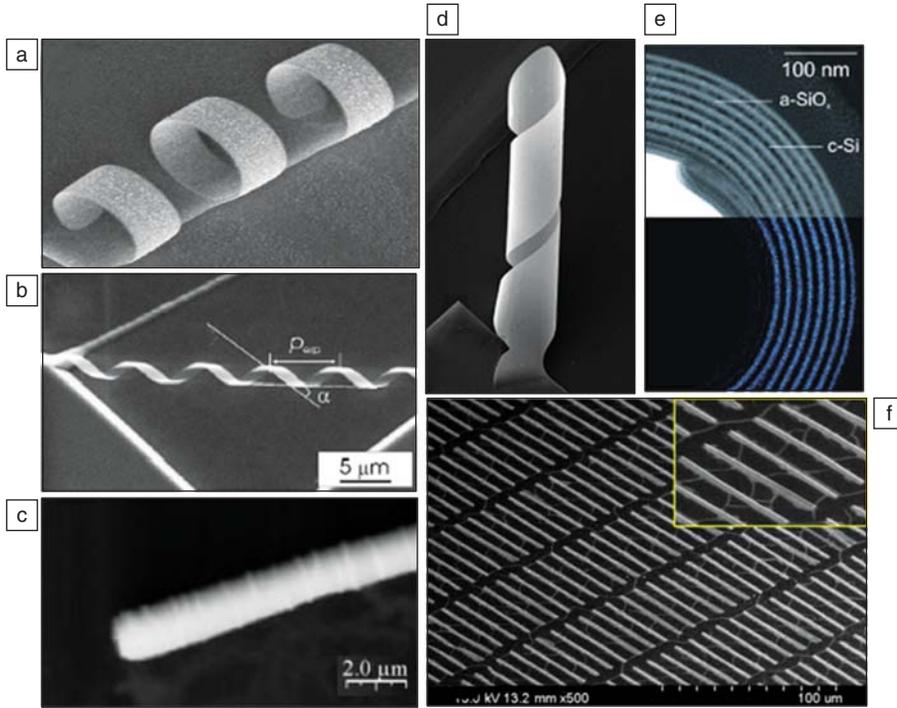


Figure 1. Examples of various types of nanoarchitectures fabricated from different materials: (a) SiGe nanorings;<sup>5</sup> (b) Si/Cr nanocoils;<sup>6</sup> (c) polymer nanotube;<sup>7</sup> (d) SiGe nanodril;<sup>5</sup> (e) cross-section of Si/SiO<sub>2</sub> multilayer nanotube;<sup>8</sup> (f) large array of GaAs/InAs nanotubes.<sup>9</sup>

that combines atomistic simulation with continuum mechanics theory has resolved discrepancies between recent experiments and classical bending theories, elucidating the difference between the mechanical response of ultrathin films and that of thick films. In addition, molecular dynamics simulations have predicted a unique self-bending mechanism of Si (or Ge) membranes for forming Si (or Ge) nanotubes and have shown the way to synthesizing carbon nanotubes with predefined size and chirality.

### Modified Timoshenko Formula

The effect of lattice misfit strain on the bending of bi- and multilayer films is described by the classical Timoshenko theory.<sup>11</sup> It has been shown that, when the film becomes very thin, the misfit strain can induce novel bending behavior not seen in thick films.<sup>1-4,12,13</sup> For example, a bilayer membrane can roll into tubular shapes of multiple rotations (like a rolled-up rug) because the radius of bending curvature becomes so small that a long membrane can roll into multiple turns;<sup>1,4</sup> roughness induced on a membrane with nanostressors (epitaxial 3D islands) can induce significant localized bending

because of the increased compliance of the ultrathin substrate;<sup>2,12</sup> or the bending curvature may exhibit an anomalous dependence on the film/substrate thickness ratio because of enhanced strain sharing.<sup>13</sup> Most existing theoretical analyses of mechanical bending of nanofilms<sup>1,4,5,14-18</sup> are performed within the framework of continuum theory, focusing on the effect of misfit strain. The classical Timoshenko formula,<sup>11</sup> for example, has been used to calculate the diameter of rolled-up nanotubes of strained bilayer films, even for films down to only a few monolayers (MLs) or a few Angstroms thick.<sup>14,15</sup> But apparent discrepancies exist between this classical theory and experimental results.<sup>15-17</sup>

Because of the large surface-to-volume ratio, the surface generally plays an important role in determining the mechanical properties of nanostructures. When the film is thinned to the nanometer scale, the effect of film atomic structure and of the intrinsic stress of the solid surface may become significant, something that is not included in the continuum theory. Therefore, the classical Timoshenko formula applies, in principle, only when the effect of surfaces can be neglected. In

bending of nanomembranes, the dominant surface effect is expected to be “surface stress.” For ultrathin films, only a few nanometers thick, surface stress due to surface reconstruction (the rearrangement of atoms on the surface from what would be a simple termination of the bulk lattice, driven by the material’s desire to reduce surface energy) or molecular adsorption can affect the film bending behavior significantly.<sup>18-20</sup>

There are two nanoscale surface stress terms: the intrinsic surface stress due to surface reconstruction (or adsorption) and the additional surface stress induced by large bending. Surface reconstruction can lead to stress because the rearrangement of surface atoms to reduce surface chemical energy can stretch or bend bonds. For the same reason, adsorption leads to stress in the surface. The surface stress of a bilayer film upon bending can be calculated as

$$\begin{aligned}\sigma_{ts} &= \sigma_{ts0} + C_{ts}\epsilon_{ts} \text{ and} \\ \sigma_{bs} &= \sigma_{bs0} + C_{bs}\epsilon_{bs},\end{aligned}\quad (1)$$

as shown in Figure 2a.  $\sigma_{ts0}$  and  $\sigma_{bs0}$  are, respectively, the intrinsic surface stresses in the top and the bottom surface.  $C_{ts}\epsilon_{ts}$  and  $C_{bs}\epsilon_{bs}$  are the bending-strain-induced additional surface stresses, where  $C_{ts}$  and  $C_{bs}$  denote the “in-plane” elastic constants of the top and bottom surface layers, and  $\epsilon_{ts}$  and  $\epsilon_{bs}$  are the strains at the top and bottom surfaces. A modified Timoshenko formula has been derived by taking into account the effects of surface stress.<sup>18</sup> For the simple case when the elastic constants for the film and substrate are the same, the modified Timoshenko formula is<sup>18</sup>

$$\begin{aligned}\kappa &= \frac{6(E_s\epsilon_m t_f)}{E_s t^2 + 6C_s' t} (1 + \beta)^{-1} \\ &+ \frac{6(C_s'\epsilon_m)}{E_s t^2 + 6C_s' t} + \frac{6(\Delta\sigma)}{E_s t^2 + 6C_s' t}.\end{aligned}\quad (2)$$

In Equation 2,  $\kappa$  is the bending curvature. The first two terms account for the effect of misfit strain and the third for the effect of intrinsic surface stress.  $E_s$  is the Young’s modulus of the substrate,  $\beta = t_f/t_s$  is the ratio of film ( $t_f$ ) and substrate thickness ( $t_s$ ),  $t = t_f + t_s$ ,  $\epsilon_m$  is the misfit strain between the film and substrate and  $\Delta\sigma = \sigma_{ts0} - \sigma_{bs0}$ .  $C_s'$  is the in-plane elastic constant of the surface layer, assuming  $C_{ts} = C_{bs} = C_s'$ . Equation 2 reduces to the classical Timoshenko formula when all the surface stress effects are absent (i.e.,  $\Delta\sigma = 0$  and  $C_s' = 0$ ). Similarly, a modified Stoney formula,<sup>21</sup> used to calculate the bending cur-

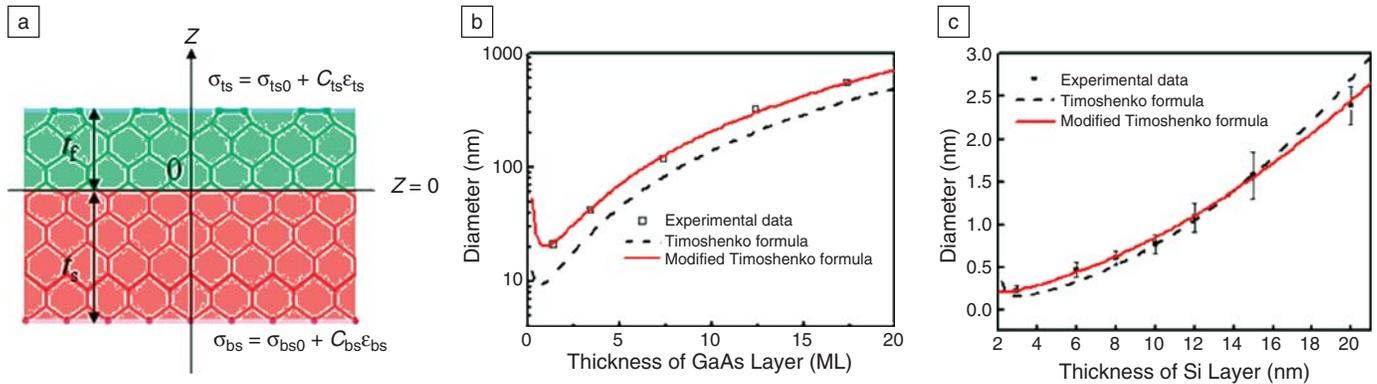


Figure 2. Comparison of results with classical and modified Timoshenko formula. (a) Physical and geometric parameters used for the derivation. (b) Diameters of GaAs/InAs nanotubes as a function of thickness of the GaAs layer with the thickness of the InAs layer fixed at two monolayers. (c) Diameters of pure Si micro- and nanotubes as a function of thickness of the Si layer with the thickness of the strained Si layer fixed at 2 nm.

vature of a single-layer film induced by an external stress load, can be derived for the bending of a single-layer film induced by adsorption on the surface.<sup>20</sup>

The modified Timoshenko formula has been confirmed by atomistic simulations<sup>18</sup> from which the bending curvatures of single-layer Si and Ge nanofilms and bilayer SiGe nanofilms are calculated. One important question is “what is the thickness at which the properties of nanofilms become different from those of thicker films?” It has been shown that for a ~10-nm Si/Ge bilayer film, the intrinsic surface stress makes the bending appreciably different from that of thicker films.<sup>19</sup> The modified Timoshenko formula has been applied to predict the diameters of GaAs/InAs and Si nanotubes.<sup>18</sup> The result gives a much better agreement with experiments than the classical Timoshenko formula, as shown respectively in Figure 2b and 2c, resolving some controversies between the theory and experiment.<sup>15–17</sup>

In modifying the classical formula, the effect of surface stress can be summarized as two main contributions. On one hand, the intrinsic surface stress substantially changes the driving force for bending beyond the effect of misfit strain, as manifested by the changed tube diameter, shown in Figure 2b for GaAs/InAs nanotubes. On the other hand, the bending-induced surface stress changes the scaling of the bending curvature with thickness, as shown in Figure 2c for Si micro- and nanotubes. Equation 2 shows that in the classical formula, the bending curvature scales with thickness as  $\kappa \sim t^{-2}$ ; in the modified formula, it is  $\kappa \sim (at^2 + bt)^{-1}$ .<sup>18,19</sup> Fitting the modified formula to the data furthermore allows an estimate of the magnitude of surface stress and surface layer elastic constants.<sup>18</sup>

### The Effect of Surface Reconstruction and Intrinsic Surface Stress

Surface stress is closely related to surface atomic structure (e.g., reconstruction, adsorption), as mentioned earlier. In this regard, atomistic simulations provide an effective tool to study the effect of surface reconstruction and hence surface stress, as illustrated in recent simulations of mechanical bending of Si/Ge nanofilms.<sup>19,20</sup> Conventionally, a solid membrane, no matter how thin, will not bend itself without the “driving force” of some externally applied stress. This is true, however, only because we have neglected the atomic details of the film structure, especially the surface atomic structure that governs the intrinsic surface stress. When the thickness is reduced to the nanometer scale, the discreteness (atomic nature) of the film structure and the effect of intrinsic surface stress become increasingly prominent. These features make the bending behavior of a nanofilm not only quantitatively but also qualitatively different from that of a macrofilm.

Consider a Si(001) nanomembrane a few atomic layers in thickness. The Si(001) surface displays a  $(2 \times 1)$  reconstruction (atomic rearrangement of surface atoms) consisting of rows of dimers to eliminate one dangling bond per surface atom (Figure 3a).<sup>22</sup> The reconstruction is termed  $(2 \times 1)$  because relative to the bulk termination, the surface unit mesh is twice as large in one direction and the same size in the other direction. As Si has the diamond structure, the surface dimers orient in two orthogonal directions (rotated by  $90^\circ$ ) in alternate atomic layers. (The alternate layers are thus said to have a  $[1 \times 2]$  reconstruction). For an even-atomic-layer-number film, the dimers on the top and

bottom surfaces are parallel to each other, as shown in Figures 3b and 3c for a six-atomic layer film; for an odd-atomic layer number film, the dimers on top and bottom are perpendicular to each other, as shown in Figure 3d for a five-atomic layer film. This odd-even alternation of surface dimer orientations has a direct impact on the bending of Si nanofilms. The  $(2 \times 1)$  reconstruction introduces a large surface stress anisotropy:<sup>22</sup> The stress is tensile along (parallel to) the dimer bond direction ( $\sigma_{\parallel}$ ) but compressive in the orthogonal (perpendicular to the dimer bond) direction ( $\sigma_{\perp}$ ). Consequently, in an even-layer number film, the intrinsic stresses on the top and bottom surface cancel each other (Figures 3b and 3c); whereas in an odd layer number film, a surface-stress imbalance exists between its top and bottom surfaces (Figure 3d), creating a unique driving force to bend the film.

Similarly, the atomic-scale surface stress imbalance may qualitatively alter the bending behavior of SiGe bilayer films. Normally, a heteroepitaxial SiGe film (Ge grown on unstrained Si is typical) always bends toward the Si side, as illustrated in Figure 3e. In the previous case of Ge grown on unstrained Si, the compressive misfit strain in the Ge layer acts as the “external” stress to bend the whole film, with the Ge tending to expand and the Si being forced to contract. (Similarly, if Ge were grown on tensilely strained Si, the Si would tend to contract, giving bending again toward Si). When the SiGe film is very thin, however, the intrinsic surface stress due to surface reconstruction also will try to bend the film. Surface stress can act either in concert with or against the misfit strain in bending the bilayer film, depending on the orientation of surface dimers. In Figure 3f, the top Si surface is

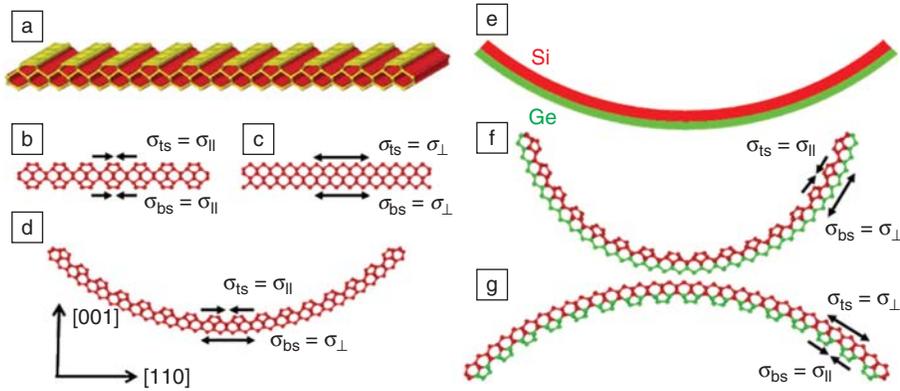


Figure 3. Effect of surface reconstruction on bending of a nanomembrane. (a) Illustration of the Si(001) ( $2 \times 1$ ) surface reconstruction, consisting of rows of dimers, which introduce a large surface stress anisotropy. (b) Side views of a six-layer Si film, showing the parallel surface dimer orientations and the balancing surface stress with the dimer bonds on both the top and bottom surfaces lying in the plane of the page. (c) Same as (b) with the dimer bonds directions on both the top and bottom surfaces into the page. (d) Side view of a five-layer Si film, showing the orthogonal surface dimer orientations and the simulated self-bending induced by surface stress imbalance. (e) Conventional picture of a SiGe bilayer film that always bends toward the Si side driven by misfit strain, because Ge has a larger lattice constant. (f) Side view of a simulated bent SiGe film demonstrating the increased bending curvature [compared to (e)] as the surface stress acts in concert with the misfit strain. (g) The surface dimers and surface stress configurations are switched as compared to (f), now opposing the misfit strain to induce a bending in the opposite direction. Arrows mark stress directions.

under tension, and the bottom Ge surface is under compression. Consequently, surface stress acts with misfit strain to increase the upward bending (toward the Si side), as seen by comparing Figure 3f to Figure 3e. However, if we switch the top and bottom surface dimer orientations, as in Figure 3g, the surface stress will act against misfit strain to decrease the upward bending. For very thin membranes, the surface stress can be so strong that it reverses the “normal” bending direction defined by misfit strain, making the film bend downward, toward the Ge side.

The atomistic simulations just discussed demonstrate that the mechanical bending of nanofilms differs qualitatively from that of macrofilms because of the dominant role played by intrinsic surface stress. In particular, ultrathin Si and Ge nanofilms that are step-free may self-bend, under their own intrinsic surface stress imbalance, arising from surface reconstruction. The same mechanism may drive SiGe bilayer nanofilms to bend toward the Ge side, opposite to what is defined by misfit strain. Of course, if either surface is stepped, the surface stress effect cancels itself averaged over the film area, but local bending may result in step-free regions, potentially leading to

surface stress-induced ripples. Similar behavior is expected also for compound-semiconductor nanofilms.

### Predicting a New Synthesis for Nanotube Formation

Another important application of atomistic simulations is to predict new synthetic processes for nanotube or microtube formation. All the original nanomechanical architectures (Figure 1) are made from bilayer or multilayer films, where each “layer” is typically at least several and often many atomic layers thick.<sup>1,4–16,23</sup> Misfit strain is employed as the only driving force for bending. The self-bending mechanism revealed by the atomistic simulations described previously leads to self-rolled-up pure Si or Ge nanotubes,<sup>19</sup> extending nanomechanical architecture to single films of one material, without the need for deposition of a second strained layer. Figure 4a shows a Si nanotube, having a radius of  $\sim 7.4$  nm, formed spontaneously from self-bending a step-free five-layer Si(001) beam.<sup>19</sup> Such a self-bending mechanism can, in principle, be applied to make nanotubes from many different materials, as long as a surface-stress imbalance can be manipulated. This capability would greatly broaden the repertoire of nanotubes for both

fundamental research and practical applications.

The surface stress-driven self-bending also modifies the formation of nanotubes from strained bilayer films when the film thickness is below the critical thicknesses at which surface effects begin to matter, mentioned in the discussion following Equation 2. Figure 4b shows a Si/Ge nanotube formed from a film of three atomic layers of Si and two atomic layers of Ge, in which surface stress dimers are oriented to have surface stress acting with the misfit film stress, as in Figure 3f.<sup>19</sup> In this case, the Si/Ge nanotube forms in the configuration it would have if there were no surface stress, but it has a much smaller radius than what would be induced by misfit strain alone. Figure 4c shows another Si/Ge nanotube, also formed from a film of three layers of Si and two layers of Ge, but it has surface stress acting against the misfit film stress, as in Figure 3g. This Si/Ge nanotube forms in a non-conventional configuration, with Ge as the inner layer, but has a larger radius, as the net stress is smaller. The self-bending mechanism found for Si and Ge films also will be applicable to III–V and other semiconductor films, where surface reconstruction typically is characterized by formation of dimer rows, creating surface stress.

Mechanisms other than surface reconstruction may create a surface stress imbalance driving nanotube formation. As suggested earlier, molecular adsorption induces surface stress. This effect has been theoretically demonstrated for synthesizing carbon nanotubes, using first principles and classical molecular dynamics simulations,<sup>24,25</sup> as shown in Figure 5 for the case of H adsorption. Single-walled nanotubes (SWNTs) are formed by realizing the process of rolling up graphene nanoribbons through adsorption of atoms at varying coverage on one side, which introduces an external stress to drive the rolling process. The method consists of three basic steps: (1) patterning of graphene nanoribbons (GNRs), (2) rolling up of GNRs into SWNTs by surface adsorption at room temperature, and (3) removing surface adsorbates by high-temperature desorption after the tubes have formed and sealed. The diameter and chirality of SWNTs can be *a priori* controlled by patterning GNRs with predefined width and direction so that the post-synthesis sorting process is eliminated. The method also potentially allows mass production of identical tubes and easy integration into device structures on a substrate. It represents a special case of applying nanomechanical architecture to

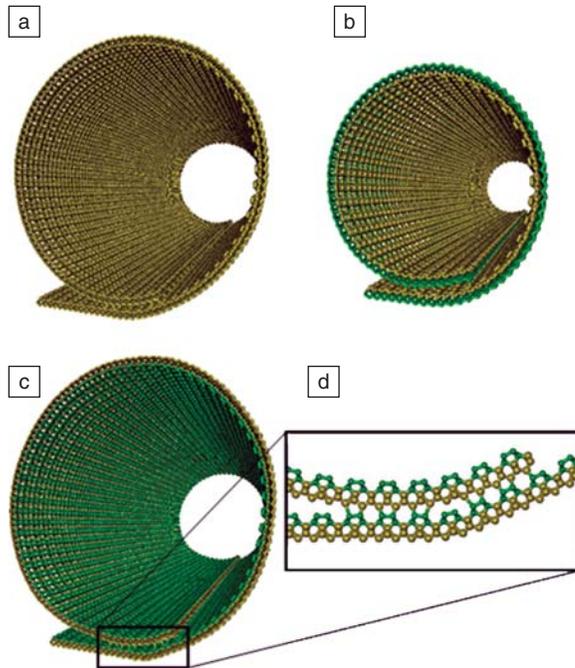


Figure 4. Nanotubes simulated by the molecular-dynamics method. (a) A Si nanotube. (b) A SiGe nanotube with Si as the inner layer. (c) A SiGe nanotube with Ge as the inner layer. (d) Enlarged view of folded edge in (c).

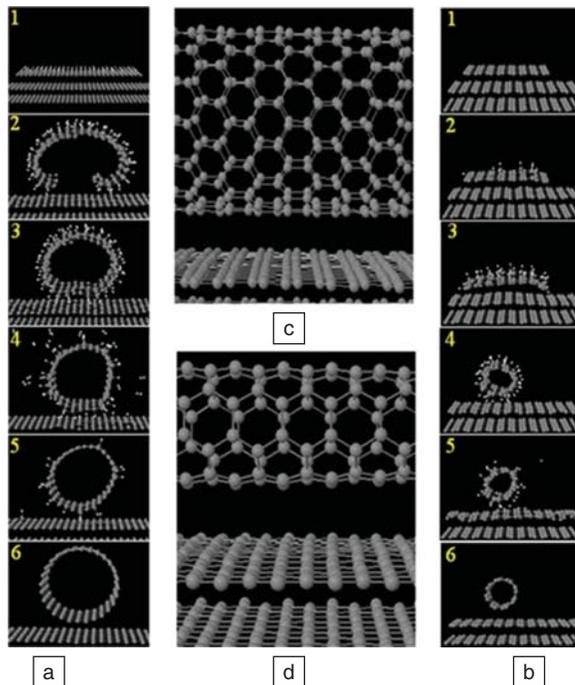


Figure 5. Classical molecular-dynamics simulation of carbon nanotube formation. (a) and (b) demonstrate the formation of 0.9 nm (diameter) zigzag and 0.5 nm (diameter) armchair single-walled nanotubes (SWNTs), respectively. (a1)–(a3) and (b1)–(b4) show the tube formation at room temperature after adsorption of H atoms. (a4)–(a6) and (b5)–(b6) show the desorption of H atoms at 1800 K. (c) and (d) show the respective side views of zigzag and armchair SWNTs. Smaller white and larger gray balls are hydrogen and carbon atoms, respectively.

the thinnest film possible, a single-atomic layer film of C.

## Conclusions

We have reviewed recent work of atomistic simulations in the context of a novel mechanics-driven nanofabrication, presenting a unique example of knowledge transfer from nanomechanical science to nanosynthesis technology. We refer the readers to recent investigations and reviews<sup>26–29</sup> on the experimental development of this approach. We believe that nanomechanical architecture will become a viable nanofabrication technique. Simulations have significantly furthered our fundamental understanding of the underlying science and offered useful guidelines for future experiments. While classical theory and continuum models provide a general description of the bending of nanomembranes induced by misfit strain, atomistic simulations are most useful in revealing the influence of surface reconstruction and adsorption on surface stress. Atomistic simulations will be an integral part of synthesizing new forms of nanostructures such as nanotubes. The simulation results on Si, Ge, SiGe, and C nanomembranes can be generalized to other nanofilms whose surface atomic structure is distinct from the underlying bulk. These findings have broad implications for nanotechnologies that employ bending of nanomembranes and films.

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- AA: Semiconductor Nanowires—  
Growth, Size-Dependent Properties, and Applications
- BB: Material Systems and Processes for Three-Dimensional Micro- and Nanoscale Fabrication and Lithography
- CC: Nanoscale Functionalization and New Discoveries in Modern Superconductivity
- DD: Ion Beams and Nano-Engineering
- EE: Materials for Nanophotonics—  
Plasmonics, Metamaterials, and Light Localization
- FF: Novel Materials and Devices for Spintronics
- GG: Electron Crystallography for Materials Research
- HH: Quantitative Characterization of Nanostructured Materials
- II: Probing Mechanics at Nanoscale Dimensions
- JJ: Nanoscale Electromechanics and Piezoresponse Force Microscopy

### SOFT MATTER, BIOLOGICAL AND BIO-INSPIRED MATERIALS

- KK: Structure-Property Relationships in Biomineralized and Biomimetic Composites
- LL: Architected Multifunctional Materials
- MM: Synthesis of Bio-inspired Hierarchical Soft and Hybrid Materials
- NN: Active Polymers
- OO: Materials and Strategies for Lab-on-a-Chip—  
Biological Analysis, Cell-Material Interfaces, and Fluidic Assembly of Nanostructures
- PP: Materials and Devices for Flexible and Stretchable Electronics

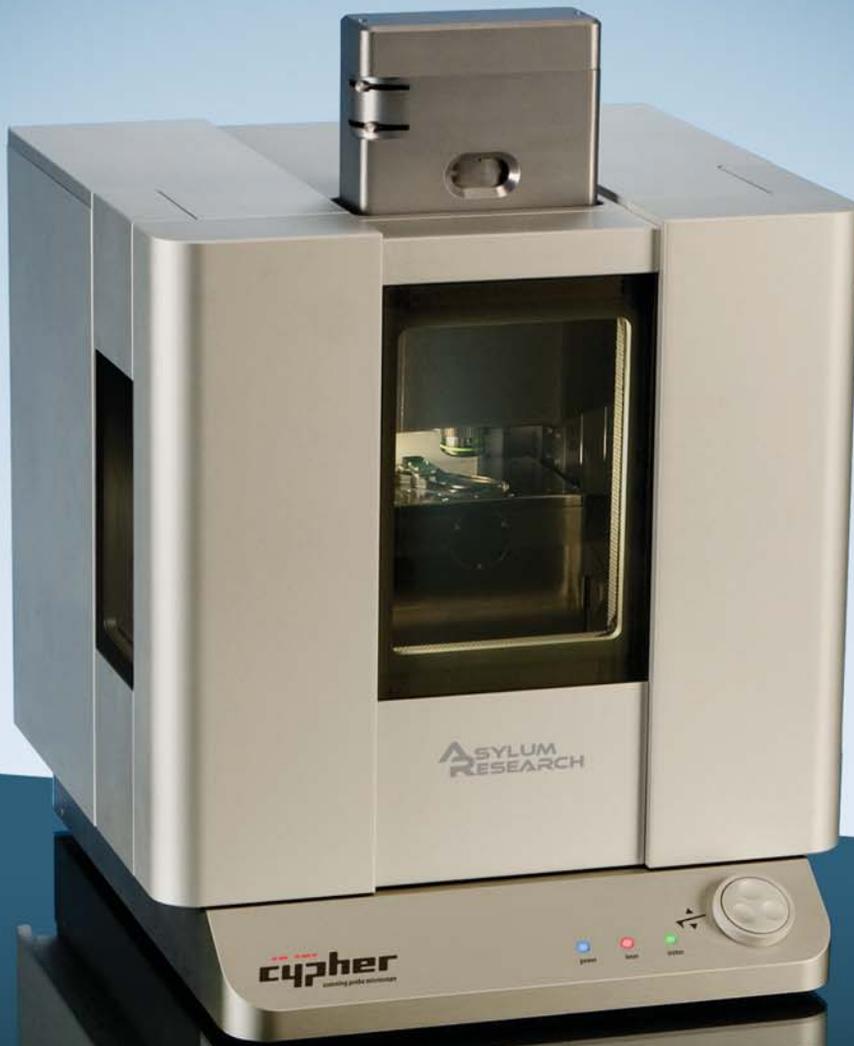
### GENERAL INTEREST

- X: Frontiers of Materials Research

# Decoding the Nanoworld

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