Modified Timoshenko formula for bending of ultrathin strained bilayer films

Ji Zang and Feng Liu[a)]
Department of Materials Science and Engineering, University of Utah, Salt Lake City, Utah 84112, USA

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Mechanical bending of nanoscale thin films can be quite different from that of macroscopic thick films. However, current understanding of mechanical bending of nanoscale thin strained bilayer films is often limited within the Timoshenko model [Timoshenko, J. Opt. Soc. Am. **11**, 233 (1925)], which was originally derived for macroscopic thick films. Here, we derive a modified Timoshenko formula by including the prominent effect of surface stress played in the nanofilms, which gives a much better agreement with the experiments than the classical formula. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2828043]

Classical bending theory was established a century ago by Stoney[1] and Timoshenko[2] in the context of bimetallic strip as used in a thermostat. The theory has since been extended for stress analysis in many different areas of applications, such as coating, epitaxial growth of thin films, and device interconnection.[3–7] Recently, with the emergence of nanotechnology, the classical bending theory has also been adopted to explain self-assembly of nanostructures via bending of strained nanoscale thin films,[8–10] even for films down to only a few monolayers (MLs, a few angstroms) thick.[11,12] Despite the fact that mechanical response of nanoscale structures, such as bending,[13,14] can be drastically different from that of macroscopic structures, most existing theoretical analyses of mechanical bending of nanofilms[8–14] are performed within the framework of continuum theory, neglecting the atomic details of film structure and the intrinsic stress of solid surface. For example, classical Timoshenko formula has been used to calculate the bending curvature (or radius) of rolled-up nanotubes of strained bilayer films. However, there exist apparent discrepancies between the theory and experimental results.[11,15,16] Here, we rederive a modified Timoshenko formula that allows us to achieve a much better agreement with experiment.

To illustrate our point, we first revisit a recent study of Si micro- and nanotubes made by releasing thin partially relaxed Si layers from their substrate by selective etching,[16] using the "nanomechanical architecture" process.[8–10] Pure Si tubes of different diameters ranging from 0.25 to 2.5 μm were fabricated from 3 to 20 nm thick Si films, as shown in Fig. 1 (square dots).

Usually, the bending curvature of a bilayer film can be analyzed using the classical Timoshenko formula[2,17] in the general form as

$$\kappa = \frac{6(E_f \varepsilon_m t_f)}{E_s t^2}\gamma, \tag{1a}$$

$$\gamma = \frac{(1+\beta)^3}{1+4\alpha\beta+6\alpha\beta^2+4\alpha\beta^3+\alpha^2\beta^4}, \tag{1b}$$

where $\alpha = E_f/E_s$, E_f and E_s are, respectively, Young's modulus of the film and substrate, $\beta = t_f/t_s$ is the ratio of film thickness t_f and substrate thickness t_s, and $t = t_f + t_s$ is the total thickness of the bilayer film. $\varepsilon_m = (a_s - a_f)/a_f$ is the misfit strain where a_s and a_f are, respectively, the lattice constant of substrate and film. For the special case of Si tubes (Fig. 1), in which both the film and substrate are made of the same material of Si (one strained and one relaxed), $\alpha = 1$ and Eq. (1) reduces to a simpler form as

$$\kappa = \frac{6\varepsilon_m t_f}{t^2}(1+\beta)^{-1}. \tag{2}$$

Equation (2) was used by Songmuang *et al.* to predict the Si tube diameters (dashed line in Fig. 1) in comparison with the experiment.[16] It was assumed that the strained (partially relaxed) Si layer (film) having a thickness $t_f = 2$ nm and under an average 2.1% tensile strain relative to the unstrained (fully relaxed) Si layer (substrate). However, such a theoretical prediction does not agree very well with the experimental results, as shown in Fig. 1.[16]

Another example is the fabrication of GaAs/InAs nanotubes from rolling up released GaAs/InAs bilayer films of only a few MLs thick.[11] Nanotubes with diameters ranging from 10 to 1000 nm were fabricated, as shown in Fig. 2 (square dots). The diameters of GaAs/InAs nanotubes have been analyzed by a couple of groups[11,15] using the classical Timoshenko formula. However, there is an apparent discrepancy between the theory (dashed line) and experiment (square dots), as shown in Fig. 2.[11] Further effort using classical Timoshenko formula but adding nonlinear and anharmonic effects[15] failed to resolve the discrepancy.

In principle, the Timoshenko formula applies only to relatively thick films for which the effect of surface on me-

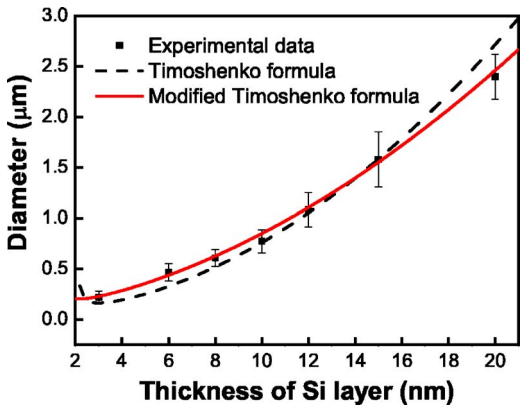

FIG. 1. (Color online) Diameters of pure Si micro- and nanotubes as a function of thickness of Si layer with fixed thickness of strained Si layer at 2 nm. See Ref. 16 for experimental details.

a)Author to whom correspondence should be addressed. Electronic mail: fliu@eng.utah.edu.

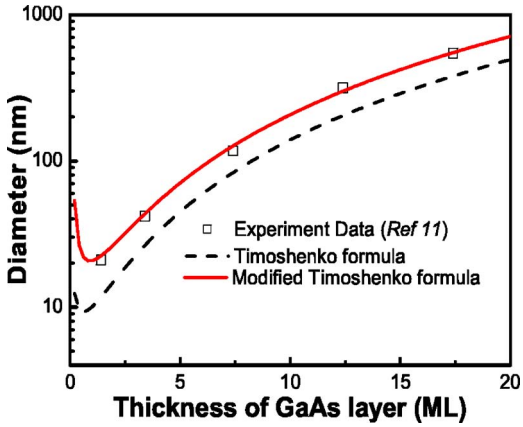

FIG. 2. (Color online) Diameters of InAs/GaAs nanotubes as a function of thickness of GaAs layer with fixed thickness of InAs layer at 2 ML. See Ref. 11 for experimental details.

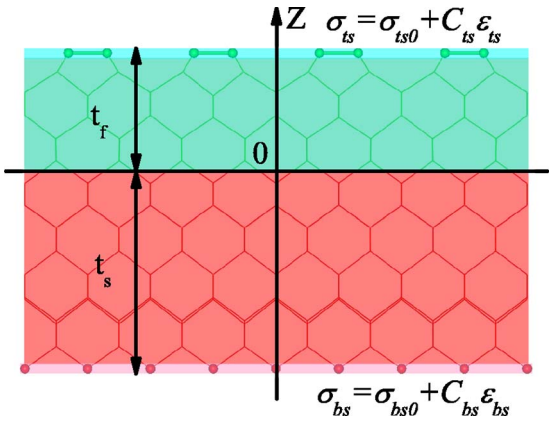

FIG. 3. (Color online) Physical and geometric parameters used for the derivation.

chanical bending can be neglected. An intrinsic property of a solid surface is its non-zero surface stress, which is generally further enhanced by surface reconstruction.[18] For ultrathin films that are only a few nanometers thick, surface stress due to surface reconstruction or molecular adsorption has been shown to affect the film bending behavior significantly.[19,20] Since misfit strain and instrinsic surface stress drive the beinding together, apparently, the Timoshenko formula that accounts only for misfit strain can not give a good description of the bending behavior of nanometer-thick bilayer films (it is not only quantitatively inaccurate but can also be qualitatively incorrect for some cases[19]). Therefore, to resolve the discrepancy between the classic theory and experimental results, as shown in Figs. 1 and 2, a modified Timoshenko formula is required for assessing the nanomechanical bending behavior of ultrathin films that are only a few nanometers thick. This has to be achieved by taking into account the effects of surface stress.

There are two nanoscale surface-stress effects need to be added: the intrinsic surface stress due to surface reconstruction and the additional surface stress induced by large bending. One usually assumes the intrinsic surface stress remains constant during and after bending. This is approximately true for a thick film of very small bending curvature. But the bending curvature increases with decreasing film thickness. For example, if a film thickness is reduced from 1 μm to 1 nm, its bending curvature will increase by six orders of magnitude. The very large bending curvature of a nanofilm means a very large bending strain in the film surface, which will in turn change the surface stress. Thus, in order to derive a more correct bending curvature formula, the bending induced additional surface stress in the top (bottom) surface must be included.

The top and bottom surface stresses of a bilayer film upon bending can be generally calculated as $\sigma_{\mathrm{ts,bs}}=\sigma_{\mathrm{ts0,bs0}}+C_{\mathrm{ts,bs}}\varepsilon_{\mathrm{ts,bs}}$. σ_{ts0} and σ_{bs0} are, respectively, the intrinsic surface stress in top and bottom surface. $C_{\mathrm{ts,bs}}\varepsilon_{\mathrm{ts,bs}}$ are the bending strain ($\varepsilon_{\mathrm{ts}}$ and $\varepsilon_{\mathrm{bs}}$) induced additional surface stress where C_{ts} and C_{bs} denote the "in-plane" elastic constants of the top and bottom surface layer, marked as the thin blue and pink layers in Fig. 3.

The bending induced strain in the top and bottom surface are, respectively, $\varepsilon_{\mathrm{ts}}=\varepsilon_m+\kappa(z_0-t_f)=\varepsilon_m+\varepsilon_a-\kappa t_s/2-\kappa t_f$, $\varepsilon_{\mathrm{bs}}=\kappa[z_0-(-t_s)]=\varepsilon_a+\kappa t_s/2$, where $\varepsilon_a=\kappa[z_0-(-t_s/2)]=\kappa z_0+\kappa t_s/2$ is introduced as the average strain of the substrate at $z=-t_s/2$. The top and bottom surface strain energies can then be calculated as (assuming $C_{\mathrm{ts}}=C_{\mathrm{bs}}=C_s'$)

$$E_{\mathrm{ts}}=A\int\sigma_{\mathrm{ts}}d\varepsilon_{\mathrm{ts}}=A\left[\sigma_{\mathrm{ts0}}\left(\varepsilon_m+\varepsilon_a-\frac{\kappa t_s}{2}-\kappa t_f\right)+\frac{C_s'}{2}\left(\varepsilon_m+\varepsilon_a-\frac{\kappa t_s}{2}-\kappa t_f\right)^2\right], \tag{3}$$

$$E_{\mathrm{bs}}=A\int\sigma_{\mathrm{bs}}d\varepsilon_{\mathrm{bs}}=A\left[\sigma_{\mathrm{bs0}}\left(\varepsilon_a+\frac{\kappa t_s}{2}\right)+\frac{C_s'}{2}\left(\varepsilon_a+\frac{\kappa t_s}{2}\right)^2\right]. \tag{4}$$

Here, A is the surface area. Now, for the case of Si tubes (Fig. 1), $\alpha=1$ for $E_f=E_s$, then the bending strain energy in the film and substrate can be calculated, respectively, as

$$E_{\mathrm{fb}}=\frac{A}{2}E_s\left(\frac{\kappa^2}{3}t_f^3+\frac{\kappa^2}{2}t_st_f^2-\kappa\varepsilon_at_f^2+\frac{\kappa^2}{4}t_s^2t_f-\kappa\varepsilon_at_st_f+\varepsilon_a^2t_f-\kappa\varepsilon_mt_f^2-\kappa\varepsilon_mt_st_f+2\varepsilon_m\varepsilon_at_f+\varepsilon_m^2t_f\right) \tag{5}$$

and

$$E_{\mathrm{sb}}=\frac{A}{2}E_s\left(\tfrac{1}{12}\kappa^2t_s^3+\varepsilon_a^2t_s\right). \tag{6}$$

Minimization of total energy $E=E_{\mathrm{ts}}+E_{\mathrm{bs}}+E_{\mathrm{fb}}+E_{\mathrm{sb}}$ with respect to κ and ε_a leads to a modified Timoshenko formula,

$$\kappa=\frac{6(E_s\varepsilon_mt_f)}{E_st^2+6C_s't}(1+\beta)^{-1}+\frac{6(C_s'\varepsilon_m)}{E_st^2+6C_s't}+\frac{6(\Delta\sigma)}{E_st^2+6C_s't}. \tag{7}$$

In Eq. (7), the first two terms account for the misfit strain effect and the third for the intrinsic surface stress effect, with $\Delta\sigma=\sigma_{\mathrm{ts0}}-\sigma_{\mathrm{bs0}}$ representing the difference of intrinsic surface stress between the top and bottom surface. It reduces to Eq. (2) when all the surface stress effects are absent, i.e., $\Delta\sigma=0$ and $C_s'=0$.

If we examine closely Fig. 1, we notice that the main difference between theory and experiment is that the two curves have a different dependence (slope) of diameters on thickness. This indicates that the classical Timoshenko formula fails to correctly predict the scaling of bending curvature with the thickness. Comparing Eqs. (7) and (2), we see that in Eq. (2) κ scales with t^{-2}, while in Eq. (7) κ scales

with $(at^2+bt)^{-1}$. This different scaling results from the bending induced surface stress contribution $(C_{ts,bs}\varepsilon_{ts,bs})$ in Eq. (7). Therefore, we will reanalyze the experimental results of pure Si tubes in Fig. 1 by including the bending induced surface stress effect. Assuming $\epsilon_m=0.021$ as in Ref. 16, we can fit the experimental data nicely using Eq. (7) with the fitting parameters $C_s'=16.45$ eV/Å^2 and $\Delta\sigma=113$ meV/Å^2 $(\sim 1.8$ J/m$^2)$. The fitted surface stress difference $(\Delta\sigma)$ is consistent with the well-known values of Si(001) surface stresss[18] as well as the typical values of the surface stress for most solid materials,[21] which are approximately 1–3 J/m^2. However, the fitted surface elastic constant C_s' seemed to be too large compared to the existing theoretical value in literature with a different sign.[22] The reason for such discrepancy needs further study. On the other hand, we note that if C_s' is set to zero, no satisfactory fitting can be obtained using either Eq. (7) by adjusting ε_m and/or $\Delta\sigma$. Thus, the physically important factor is that the diameters of nanotubes scales with film thickness differently due to the atomic-level surface stress effects, which cannot be predicted by the classical bending theory with or without nonlinear and anharmonic effects.[15]

Next, we turn our attention to the experimental data of GaAs/InAs nanotubes in Fig. 2. We notice that the main difference between the experimental data (square dots) and theoretical prediction by classical Timoshenko formula (dashed line) is an almost constant shift of curves. We expect this shift is caused by the difference of intrinsic surface stress between the top GaAs surface and bottom InAs surface. Whereas the effect of bending induced surface stress, which will affect the slope of the bending curve as discussed above for Si nanotubes, is less important and negligible. Thus, setting $C_{ts}=C_{bs}=0$ and using $\alpha\neq 1$, we derived the following modified Timoshenko formula for this special case:

$$\kappa=\frac{6(E_f\varepsilon_m t_f)+6(G_1\sigma_{ts0}-G_2\sigma_{bs0})}{E_s t^2}\gamma \tag{8a}$$

$$G_1=\frac{1+2\beta+\alpha\beta^2}{1+\beta},\quad G_2=\frac{1+2\alpha\beta+\alpha\beta^2}{1+\beta}. \tag{8b}$$

Equation (8) gives an excellent agreement with experiment shown in Fig. 2 as solid red line with $\sigma_{ts0}=49.1$ meV/Å^2 and $\sigma_{bs0}=54.9$ meV/Å^2. Again, the fitted surface stresses are consistent with typical values of solid surfaces.[21]

For the most general cases, we will have different elastic constants in the film and substrate $(E_f\neq E_s)$, different intrinsic surface stress $(\sigma_{ts0}\neq\sigma_{bs0})$ and different in-plane elastic constants $(C_{ts}\neq C_{bs})$ in top and bottom surfaces. Then we must use the most general form of the modified Timoshenko formula derived as the following:

$$\kappa=\frac{6(E_f\varepsilon_m t_f)F+6(G_1\sigma_{ts0}-G_2\sigma_{bs0})}{E_s t^2+4t(H_1C_{ts}+H_2C_{bs})+12C_{ts}C_{bs}E_s^{-1}(1+\beta)\gamma}\gamma, \tag{9a}$$

$$F=1+\frac{C_{bs}(2+\beta)}{E_s t}+\frac{2C_{ts}C_{bs}(1+\beta)+C_{ts}E_s t(1+2\beta)/(1+\beta)}{E_s E_f t t_f}, \tag{9b}$$

$$G_1=\frac{1+2\beta+\alpha\beta^2}{1+\beta}+\frac{2(1+\beta)C_{bs}}{E_s t},$$

$$G_2=\frac{1+2\alpha\beta+\alpha\beta^2}{1+\beta}+\frac{2(1+\beta)C_{ts}}{E_s t}, \tag{9c}$$

$$H_1=(1+\beta)^{-2}(1+3\beta+3\beta^2+\alpha\beta^3)\gamma,$$

$$H_2=(1+\beta)^{-2}(1+3\alpha\beta+3\alpha\beta^2+\alpha\beta^3)\gamma. \tag{9d}$$

This general formula has been shown to agree very well with molecular dynamics simulation results of Si/Ge nanotubes.[19]

In conclusion, we have derived the modified Timoshenko formula for calculating the bending curvature of nanoscale strained bilayer films using an energy minimization scheme within the framework of continuum mechanics. The main modification over the classical formula is to include the prominent role of surface stress in two important manifestations. One is the effect of intrinsic surface stress due to difference in surface structure and morphology, which changes the bending magnitude. The other is the effect of bending induced surface stress due to the large bending curvature, which changes the scaling of bending curvature with thickness. The modified Timoshenko formula has been used to predict the diameters of Si and GaAs/InAs nanotubes made from folding of strained bilayer nanofilms, giving a much better agreement with experiments.

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