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Wrinkling of a stiff thin film bonded to a pre-strained, compliant substrate with finite thickness

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A stiff thin film bonded to a pre-strained, compliant substrate wrinkles into a sinusoidal form upon release of the pre-strain. Many analytical models developed for the critical pre-strain for wrinkling assume that the substrate is semi-infinite. This critical pre-strain is actually much smaller than that for a substrate with finite thickness (Ma Y *et al.* 2016 *Adv. Funct. Mater.* (doi:10.1002/adfm.201600713)). An analytical solution of the critical pre-strain for a system of a stiff film bonded to a pre-strained, finite-thickness, compliant substrate is obtained, and it agrees well with the finite-element analysis. The finite-thickness effect is significant when the substrate tensile stiffness cannot overwhelm the film tensile stiffness.

1. Introduction

A stiff film bonded to a pre-strained, compliant substrate wrinkles upon releasing the pre-strain [1,2]. Such a system has many important applications in stretchable

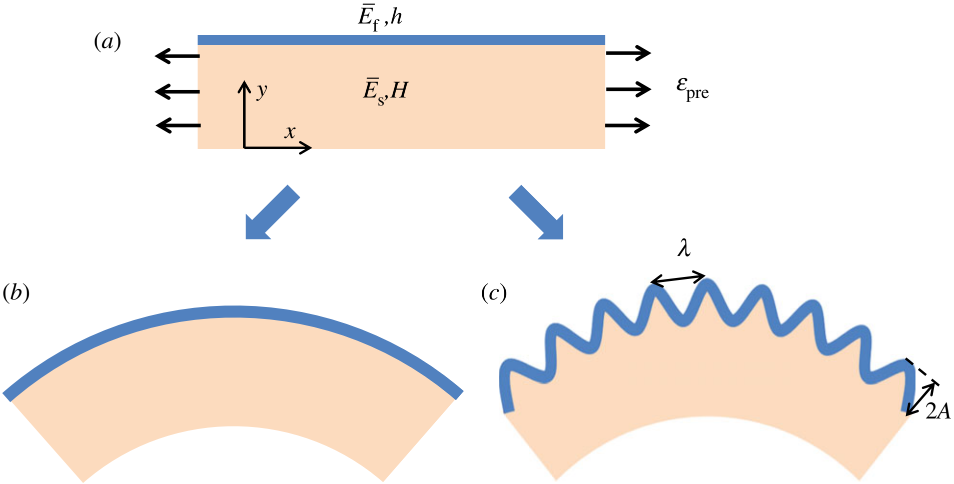

Figure 1. Schematic illustrations. (*a*) A stiff thin film bonded to a pre-strained, compliant substrate with finite thickness; (*b*) bending of the film/substrate system upon release of the small pre-strain; and (*c*) wrinkling of the stiff thin film, along with bending of the film/substrate system, upon release of large pre-strain. (Online version in colour.)

inorganic electronics [3–8], micro/nano pattern formation [9–11], high-precision micro/nano measurement techniques [12], tuneable metamaterials [13], nanocomposites [14], stretchable transistors [15] and biomimetic materials [16]. Analytical models have been developed for wrinkling of a stiff thin film on a pre-strained compliant substrate [17–21]. The results identify the critical pre-strain for wrinkling, below which the film remains flat. However, all of these studies assume that the substrate is semi-infinite such that its tensile stiffness overwhelms that of the film. Consequently, the substrate recovers the initial length after the pre-strain is released and its bottom remains flat.[1]

The critical pre-strain for wrinkling obtained for a semi-infinite substrate, however, is smaller than the numerical and experimental results for a substrate with finite thickness [22], even for substrates that are more than 1000 times thicker than the film. This is because the substrate elastic modulus E_s is often more than five orders of magnitude smaller than the film elastic modulus E_f [1,2], such that the substrate tensile stiffness E_sH cannot overwhelm the film tensile stiffness E_fh, where H and h are the substrate and film thicknesses, respectively (figure 1*a*). Consequently,

(1) the substrate cannot shrink back to its initial length after release of the pre-strain; and
(2) the film/substrate system may bend after the pre-strain is released (figure 1*b*).

The recent study by Ma *et al*. [22] accounted for (1), while this paper aims to establish an analytic model for both (1) and (2). The resulting critical pre-strain will be useful for many applications such as the strain-limiting design of materials [22] and tuneable optical design of the intensity for diffraction peaks [23].

2. Analytical model

A stiff thin film is bonded onto a pre-strained (ε_{pre}), compliant substrate (figure 1*a*). For small pre-strain, the stiff film does not wrinkle upon release of the pre-strain; instead, the film and substrate bend (figure 1*b*). Let ε denote the membrane strain in the film. The strain in the substrate

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[1] Huang *et al*. [24] studied the finite thickness of a substrate subjected to compression, to be discussed below.

is $\varepsilon_s(y) = \varepsilon_{pre} + \varepsilon - \kappa(H - y)$, where κ is the curvature of the substrate, and the co-ordinate y is shown in figure 1*a*. The potential energy is

$$U_{bend} = \frac{1}{2}\bar{E}_f h\varepsilon^2 + \frac{1}{2}\bar{E}_s \int_0^H [\varepsilon_{pre} + \varepsilon - \kappa(H - y)]^2 dy, \tag{2.1}$$

where $\bar{E}_f = E_f/(1 - \upsilon_f^2)$ and $\bar{E}_s = E_s/(1 - \upsilon_s^2)$ are the plane-strain moduli of the stiff thin film and compliant substrate, respectively, and υ_f and υ_s are the Poisson's ratios.

Minimization of the potential energy $\partial U_{bend}/\partial\varepsilon = 0$ and $\partial U_{bend}/\partial\kappa = 0$ gives $\varepsilon = -\bar{E}_s H\varepsilon_{pre}/(4\bar{E}_f h + \bar{E}_s H)$ and $\kappa = -6\bar{E}_f h\varepsilon_{pre}/[H(4\bar{E}_f h + \bar{E}_s H)]$. Equation (2.1) then becomes

$$U_{bend} = \frac{\bar{E}_f h\bar{E}_s H\varepsilon_{pre}^2}{2(4\bar{E}_f h + \bar{E}_s H)}. \tag{2.2}$$

Once the pre-strain exceeds the critical pre-strain (to be determined), the stiff film wrinkles on the top surface of the substrate (figure 1*c*) and the film/substrate bends. In addition to the membrane strain ε, the film is also subjected to wrinkling with amplitude A and period λ to be determined. The strain energy in the film is [24]

$$U_{film} = \frac{1}{2}\bar{E}_f h\varepsilon^2 - \frac{1}{4}\bar{E}_f h|\varepsilon|k^2A^2 + \frac{1}{32}\bar{E}_f hk^4A^4 + \frac{1}{48}\bar{E}_f h^3k^4A^2, \tag{2.3}$$

which degenerates to the first term on the right-hand side of equation (2.1) when the amplitude $A = 0$; here $k = 2\pi/\lambda$. The strain energy in the substrate is

$$U_{substrate} = \frac{1}{2}\bar{E}_s \int_0^H [\varepsilon_{pre} + \varepsilon - \kappa(H - y)]^2 dy + \frac{\bar{E}_s}{4}kA^2 g(kH), \tag{2.4}$$

which degenerates to the last term in equation (2.1) when the amplitude $A = 0$. The last term in the above equation is the strain energy in the substrate due to wrinkling [24], and the function g is

$$g(x) = \frac{\cosh(2x) + 1 + 2x^2}{2\sinh(2x) - 4x}, \tag{2.5}$$

for an incompressible substrate ($\upsilon_s = 0.5$), and g approaches 1/2 for a semi-infinite substrate. The potential energy is the sum of U_{film} and $U_{substrate}$,

$$\begin{aligned} U_{bend+wrinkle} =& \frac{1}{2}\bar{E}_f h\varepsilon^2 + \frac{1}{2}\bar{E}_s \int_0^H [\varepsilon_{pre} + \varepsilon - \kappa(H - y)]^2 dy \\ &+ \frac{1}{4}\bar{E}_f h(f - |\varepsilon|)k^2A^2 + \frac{1}{32}\bar{E}_f hk^4A^4, \end{aligned} \tag{2.6}$$

where

$$f = \frac{k^2h^2}{12} + \frac{\bar{E}_s g(kH)}{kh\bar{E}_f}. \tag{2.7}$$

Minimization of the potential energy with respect to k and A, $\partial U_{bend+wrinkle}/\partial k = 0$ and $\partial U_{bend+wrinkle}/\partial A = 0$, gives

$$6\frac{g(kH) - g'(kH)kH}{(kH)^3} = \frac{\bar{E}_f h^3}{\bar{E}_s H^3} \tag{2.8}$$

and

$$k^2A^2 = 4(|\varepsilon| - f), \tag{2.9}$$

where $g'(x) = dg(x)/dx$. Equation (2.8) suggests that the normalized period, $\lambda/[(\bar{E}_f/\bar{E}_s)^{1/3}h]$, or equivalently $kh/(\bar{E}_s/\bar{E}_f)^{1/3}$, depends only on the film/substrate bending stiffness ratio $\bar{E}_f h^3/\bar{E}_s H^3$, as shown in figure 2. The period becomes independent of the substrate thickness H when the bending stiffness ratio $\bar{E}_f h^3/\bar{E}_s H^3$ is less than 0.01, which is consistent with Huang *et al.* [24].

Minimization of the potential energy with respect to ε and κ, $\partial U_{bend+wrinkle}/\partial\varepsilon = 0$ and $\partial U_{bend+wrinkle}/\partial\kappa = 0$, gives $\varepsilon = 4\bar{E}_f hf/(\bar{E}_s H) - \varepsilon_{pre}$, and $\kappa = 6\bar{E}_f hf/(\bar{E}_s H^2)$, where f is obtained

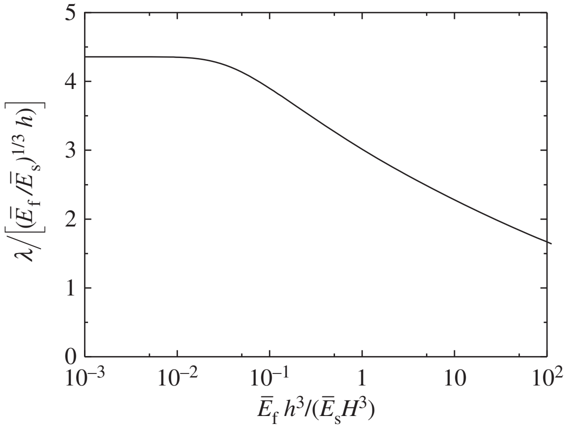

Figure 2. The normalized wrinkle period $\lambda/[(\bar{E}_f/\bar{E}_s)^{1/3}h]$ versus the film-to-substrate bending stiffness ratio $[\bar{E}_f h^3/(\bar{E}_s H^3)]$.

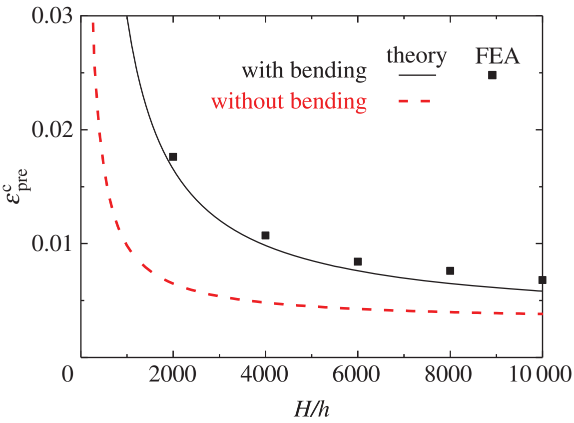

Figure 3. The critical pre-strain $\varepsilon^{\text{c}}_{\text{pre}}$ versus the substrate-to-film thickness ratio (H/h) for a polyimide film on a PDMS substrate. FEA, finite-element analysis; PDMS, polydimethylsiloxane. (Online version in colour.)

from equation (2.7). The potential energy then becomes

$$U_{\text{bend+wrinkle}} = \bar{E}_f h f \left[\varepsilon_{\text{pre}} - \frac{1}{2}\left(\frac{4\bar{E}_f h}{\bar{E}_s H} + 1\right) f \right]. \tag{2.10}$$

Comparison of the potential energy in equations (2.2) and (2.10) suggests that wrinkling occurs when $U_{\text{bend}} > U_{\text{bend+wrinkle}}$, which gives

$$\varepsilon_{\text{pre}} > \left(\frac{4\bar{E}_f h}{\bar{E}_s H} + 1\right) f = \left(\frac{4\bar{E}_f h}{\bar{E}_s H} + 1\right)\left[\frac{k^2 h^2}{12} + \frac{\bar{E}_s g(kH)}{kh\bar{E}_f}\right], \tag{2.11}$$

where k, f and g are obtained from equations (2.5), (2.7) and (2.8), respectively. It should be pointed out that equation (2.11) also ensures that the right-hand side of equation (2.9) is positive such that there is a solution for the amplitude A.

3. Discussion

When the bending stiffness of the substrate overwhelms that of the film, i.e. $\bar{E}_{\mathrm{f}}h^3/(\bar{E}_{\mathrm{s}}H^3) <\sim 0.01$, equation (2.11) can be further simplified as

$$\varepsilon_{\mathrm{pre}} > \varepsilon_{\mathrm{pre}}^{\mathrm{c}} = \frac{1}{4}\left(\frac{4\bar{E}_{\mathrm{f}}h}{\bar{E}_{\mathrm{s}}H}+1\right)\left(\frac{3\bar{E}_{\mathrm{s}}}{\bar{E}_{\mathrm{f}}}\right)^{2/3}. \tag{3.1}$$

For $H \to \infty$, the above equation degenerates to that for a semi-infinite substrate [1,2]. The critical pre-strain $\varepsilon_{\mathrm{pre}}^{\mathrm{c}}$ in equation (3.1) is larger than $\frac{1}{4}((\bar{E}_{\mathrm{f}}h/\bar{E}_{\mathrm{s}}H)+1)(3\bar{E}_{\mathrm{s}}/\bar{E}_{\mathrm{f}})^{2/3}$ [22], which neglects the effect of film/substrate bending. Figure 3 shows the critical pre-strain $\varepsilon_{\mathrm{pre}}^{\mathrm{c}}$ versus the thickness ratio H/h for a polyimide film ($E_{\mathrm{f}} = 2.5\,\mathrm{GPa}$, $\nu_{\mathrm{f}} = 0.34$) on a polydimethylsiloxane substrate ($E_{\mathrm{s}} = 1\,\mathrm{MPa}$, $\nu_{\mathrm{s}} = 0.5$). The results obtained from finite-element analysis agree well with the critical pre-strain in equation (3.1).

Competing interests. We declare we have no competing interests.

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References

1. Bowden N, Brittain S, Evans AG, Hutchinson JW, Whitesides GM. 1998 Spontaneous formation of ordered structures in thin films of metals supported on an elastomeric polymer. *Nature* **393**, 146–149. (doi:10.1038/30193)
2. Khang DY, Jiang HQ, Huang Y, Rogers JA. 2006 A stretchable form of single-crystal silicon for high-performance electronics on rubber substrates. *Science* **311**, 208–212. (doi:10.1126/science.1121401)
3. Ko HC *et al.* 2009 Curvilinear electronics formed using silicon membrane circuits and elastomeric transfer elements. *Small* **5**, 2703–2709. (doi:10.1002/smll.200900934)
4. Sun YG, Kumar V, Adesida I, Rogers JA. 2006 Buckled and wavy ribbons of GaAs for high-performance electronics on elastomeric substrates. *Adv. Mater.* **18**, 2857–2862. (doi:10.1002/adma.200600646)
5. Sun YG, Choi WM, Jiang HQ, Huang YGY, Rogers JA. 2006 Controlled buckling of semiconductor nanoribbons for stretchable electronics. *Nat. Nanotechnol.* **1**, 201–207. (doi:10.1038/nnano.2006.131)
6. Kaltenbrunner M *et al.* 2013 An ultra-lightweight design for imperceptible plastic electronics. *Nature* **499**, 458–463. (doi:10.1038/nature12314)
7. Xu F, Zhu Y. 2012 Highly conductive and stretchable silver nanowire conductors. *Adv. Mater.* **24**, 5117–5122. (doi:10.1002/adma.201201886)
8. Oyewole OK, Yu D, Du J, Asare J, Oyewole DO, Anye VC, Fashina A, Kana MGZ, Soboyejo WO. 2015 Micro-wrinkling and delamination-induced buckling of stretchable electronic structures. *J. Appl. Phys.* **117**, 235501. (doi:10.1063/1.4922665)
9. Moon MW, Lee SH, Sun JY, Oh KH, Vaziri A, Hutchinson JW. 2007 Wrinkled hard skins on polymers created by focused ion beam. *Proc. Natl Acad. Sci. USA* **104**, 1130–1133. (doi:10.1073/pnas.0610654104)
10. Tang J, Guo H, Chen M, Yang PT, Tsoukalas D, Zhang BZ, Liu J, Xue CY, Zhang WD. 2015 Wrinkled Ag nanostructured gratings towards single molecule detection by ultrahigh surface Raman scattering enhancement. *Sensor Actuat. B Chem.* **218**, 145–151. (doi:10.1016/j.snb.2015.04.008)
11. Sun J-Y, Xia SM, Moon M-W, Oh KH, Kim K-S. 2012 Folding wrinkles of a thin stiff layer on a soft substrate. *Proc. R. Soc. A* **468**, 932–953. (doi:10.1098/rspa.2011.0567)
12. Wilder EA, Guo S, Lin-Gibson S, Fasolka MJ, Stafford CM. 2006 Measuring the modulus of soft polymer networks via a buckling-based metrology. *Macromolecules* **39**, 4138–4143. (doi:10.1021/ma060266b)

13. Lee S, Kim S, Kim TT, Kim Y, Choi M, Lee SH, Kim JY, Min B. 2012 Reversibly stretchable and tunable terahertz metamaterials with wrinkled layouts. *Adv. Mater.* **24**, 3491–3497. (doi:10.1002/adma.201200419)
14. Semler MR, Harris JM, Hobbie EK. 2014 Wrinkling and folding of nanotube-polymer bilayers. *J. Chem. Phys.* **141**, 044901. (doi:10.1063/1.4887775)
15. Wu MY, Zhao J, Xu F, Chang TH, Jacobberger RM, Ma ZQ, Arnold MS. 2015 Highly stretchable carbon nanotube transistors enabled by buckled ion gel gate dielectrics. *Appl. Phys. Lett.* **107**, 053301. (doi:10.1063/1.4928041)
16. Yan D, Zhang K, Hu GK. 2016 Wrinkling of structured thin films via contrasted materials. *Soft Matter* **12**, 3937–3942. (doi:10.1039/c6sm00228e)
17. Jiang HQ, Khang DY, Fei HY, Kim H, Huang YG, Xiao JL, Rogers JA. 2008 Finite width effect of thin-films buckling on compliant substrate: experimental and theoretical studies. *J. Mech. Phys. Solids.* **56**, 2585–2598. (doi:10.1016/j.jmps.2008.03.005)
18. Chen X, Hutchinson JW. 2004 Herringbone buckling patterns of compressed thin films on compliant substrates. *J. Appl. Mech. ASME* **71**, 597–603. (doi:10.1115/1.1756141)
19. Song J, Jiang H, Liu ZJ, Khang DY, Huang Y, Rogers JA, Lu C, Koh CG. 2008 Buckling of a stiff thin film on a compliant substrate in large deformation. *Int. J. Solids. Struct.* **45**, 3107–3121. (doi:10.1016/j.ijsolstr.2008.01.023)
20. Jiang HQ, Khang DY, Song JZ, Sun YG, Huang YG, Rogers JA. 2007 Finite deformation mechanics in buckled thin films on compliant supports. *Proc. Natl Acad. Sci. USA* **104**, 15 607–15 612. (doi:10.1073/pnas.0702927104)
21. Huang R, Suo Z. 2002 Wrinkling of a compressed elastic film on a viscous layer. *J. Appl. Phys.* **91**, 1135–1142. (doi:10.1063/1.1427407)
22. Ma Y *et al.* 2016 Design of strain-limiting substrate materials for stretchable and flexible electronics. *Adv. Funct. Mater*. (doi:10.1002/adfm.201600713)
23. Harrison C, Stafford CM, Zhang WH, Karim A. 2004 Sinusoidal phase grating created by a tunably buckled surface. *Appl. Phys. Lett.* **85**, 4016–4018. (doi:10.1063/1.1809281)
24. Huang ZY, Hong W, Suo Z. 2005 Nonlinear analyses of wrinkles in a film bonded to a compliant substrate. *J. Mech. Phys. Solids* **53**, 2101–2118. (doi:10.1016/j.jmps.2005.03.007)