WRINKLING MECHANICS OF COMPLIANT MATERIALS AND FILM/SUBSTRATE SYSTEMS WITH EMPHASIS ON SUBSTRATE NONLINEARITY

These slides are drawn from (2011-5), (2012-1), (2012-2) & (2012-3) & 1 paper to be published

Wrinkling of neo-Hookean film on infinitely deep neo-Hookean substrate

An exact finite strain analysis for bifurcation under plane strain compression

\[ \varepsilon = 1 - \lambda \]

nominal compressive strain at wrinkling

Finite amplitude creases can form

Transition from stable to unstable wrinkling

Stiff film on deep compliant substrate
Highly stable and imperfection-insensitive

Biot’s surface wrinkling of half-space
Highly unstable and imperfection sensitive

Ratio of film and substrate shear moduli \( \mu_f / \mu_S \)
Crease, or sulcus, structure on surface of the brain from Hohlfeld & Mahadevan (2011)

Crease on the surface on a starch-gel from Hong, Zhao and Suo (2009).

Without stiff film, wrinkles are generally not observed—creases form

Wrinkled wrinkles

Wrinkled crust of earth on the plane of Nambia (from K.S. Kim)

With a stiff film, wrinkles are observed and highly stable

50nm gold film on PDMS
Plane Strain Compression of neo-Hookean Film on Neo-Hookean Substrate

--stiff film wrinkling
--substrate wrinkling with no film
--substrate creasing with no film
--stability of bifurcation as dependent on film/substrate elastic mismatch

**Nonlinear wrinkling behavior of a stiff film on compliant substrate in plane strain** (2004-7)

Buckling amplitude

\[ \varepsilon_W = \frac{1}{4} \left( \frac{3 \mu_s}{\mu_f} \right)^{1/3} \]

Overall nominal compressive strain

\[ \varepsilon = 1 - \lambda \]

Highly stable bifurcation behavior for compressive strains 10 or 20 times the bifurcation strain

A single bifurcation mode. Deformation decays exponentially into substrate with length \( \ell \)

\[ \ell = \frac{2 \pi h}{\left( \frac{3 \mu_s}{\mu_f} \right)^{2/3}} \]

Normal deflection of film:

\[ w = \xi h \cos \left( x / \ell \right), \quad \xi = \sqrt{\frac{\varepsilon}{\varepsilon_W}} - 1 \]

This is an exact finite amplitude solution for an elastic film (von Karman plate) on a *linear* elastic substrate. Note that the bifurcation strain is typically on the order of 1% or less for stiff films. Nonlinear substrate effects do not become important until overall strains are on the order of 20% when period-doubling occurs.

50 nm silica film on PDMS
Unstable bifurcation behavior and extreme imperfection-sensitivity of wrinkling of a neo-Hookean substrate with no film (2012-1)

Biot’s critical wrinkling strain: $\varepsilon_w = 1 - \lambda_w = 0.456$

Multiple modes:
$$w(x_1, x_2) = \xi \ell \cos\left(\frac{2\pi x_1}{\ell}\right) f\left(\frac{x_2}{\ell}\right)$$
for any $\ell$!

$$f(x_2 / \ell) \to 0, \quad x_2 / \ell \to -\infty$$

Mode shape is an incipient crease.
Due to nonlinear interaction among critical modes wrinkling is highly unstable and extremely imperfection-sensitive.

Initial imperfection: an initial sinusoidal undulation of the surface in the shape of mode 1 in the unloaded state:
$$\delta(x_1) = \bar{\xi}_1 \ell \cos\left(\frac{2\pi x_1}{\ell}\right)$$

Asymptotic result for the max compressive strain at wrinkling instability:
$$\varepsilon_w - \varepsilon^* = 1.37\sqrt{\bar{\xi}_1}$$

An undulation amplitude of 1% of its wavelength reduces the wrinkling strain by 1/3.
Crease instabilities (*Non-bifurcation modes*):
Finite strain modes that can exist below the Biot bifurcation strain

Recall Biot’s wrinkling bifurcation strain

\[ \varepsilon_w = 0.456 \]

Finite strain amplitude crease (sulcus) modes exist at compressive strains above

\[ \varepsilon_{\text{CREASE}} = 0.35 \]

Like the Biot wrinkle, the crease size is indeterminant—it can be arbitrarily small.

Hohlfeld (2008), Hong, Zhao & Suo (2009), Hohlfeld & Mahavaden (2011)
Results from Y. Cao’s numerical analysis of periodic solutions (2012-1):

Strains at the onset of wrinkling instability with sinusoidal imperfection

\[
\delta(x_1) = \overline{\xi}_1 l \cos \left( \frac{2\pi x_1}{l} \right)
\]

imperfection

Wrinkle become unstable when local strain at A reaches wrinkling strain:

\[
\epsilon_{\text{WRINKLE}} = 0.456
\]

And a crease abruptly forms as described on next slide.
Transition from stable to unstable bifurcation for thin film on substrate (neo-Hookean materials)

An exact Koiter analysis (Hutch, paper to be published)

Normal deflection of film: \( w = \xi h \cos(x/l) \)

Initial post-bifurcation expansion: \( \varepsilon / \varepsilon_C = 1 + b \xi^2 \)

Pre-stretch of substrate: \( r_S^0 = \lambda_2^0 / \lambda_1^0 = 1 / \lambda_1^{02} \lambda_3^0 \)

No pre-stretch of substrate:
Digression—Fundamental issues in bifurcation and stability (Koiter, 1945)

Pre-buckling solution
\[ u = u_0(L) + \tilde{u}, \quad P(u, \dot{L}) = P_2(\tilde{u}, L) + P_3(\tilde{u}, L) + P_4(\tilde{u}, L) + \ldots \]

Potential energy functional
Additional displacement
Load parameter
Buckling mode (eigenmode)
Buckling load (eigenvalue)

\[ P_2(\tilde{u}, L) \text{ is the quadratic bifurcation functional giving } \tilde{u}_C \text{ & } L_C \]

**Stability condition:** \[ P_2(\tilde{u}, L) > 0 \Rightarrow P(\tilde{u}, L) > 0 \text{ for all } |\tilde{u}| < \varepsilon \]

True for all discrete (finite dimensional) systems and, for example, for continuum plate & shell theories. BUT, this is not true for nonlinear elastic solids (e.g., for a neo-Hookean solid)

Wrinkling bifurcation (Biot, 1963)
\[ P_2(\tilde{u}_C, L_C) = 0 \Rightarrow \varepsilon_{Wrinkle} = 0.45 \]

Crease instability (Holfeld & Mahavaden, 2011)
\[ P_2(\tilde{u}_{Crease}, L_{Crease}) > 0, \text{ but } \varepsilon_{Crease} = 0.35 \]

Neo-Hookean substrate subject to plane strain compression

The wavelength of the wrinkle and the scale of the crease are arbitrary.

There is no length scale in the problem
Continuing digression: Why does classical buckling (bifurcation) theory break down for nonlinear continuum elasticity?

Plates: \[ P_2(\tilde{w}, L) = \frac{1}{2} \int_S \left( D (\nabla^2 \tilde{w})^2 + L \left( N_{\alpha\beta}^0 \tilde{w}_\alpha \tilde{w}_\beta \right) \right) dS \]

For the crease

Neo-Hookean elasticity: \[ P_2(\tilde{u}, L) = \frac{\mu}{2} \int_S \left( (\tilde{u}_{1,1}^2 + \tilde{u}_{2,2}^2 + \tilde{u}_{1,2}^2 + \tilde{u}_{2,1}^2) - L (\tilde{u}_{1,1}\tilde{u}_{2,2} - \tilde{u}_{1,2}\tilde{u}_{2,1}) \right) dS \]

\[ (L\tilde{u}_{1,1} + \tilde{u}_{2,2} = 0) \]

\[ u = u_0(L) + \tilde{u}, \quad P(u, L) = P_2(\tilde{u}, L) + P_3(\tilde{u}, L) + P_4(\tilde{u}, L) + \ldots \]

\[ P_3(\tilde{u}, L) \sim \mu \int_S (\tilde{u}_{1,1}^3 + \ldots) dS \]

Classical buckling problems (e.g., plates & shells) have a definite wavelength.

The wavelength of wrinkles & creases can be arbitrarily small.

The crease is a finite strain mode (not a bifurcation mode) for which it is possible to have

\[ |P_2(\tilde{u}, L)| < |P_3(\tilde{u}, L)| \quad \text{as} \quad |\tilde{u}| \to 0 \]

as the size of the crease goes to zero

End of digression
Y. Cao's simulations for plane strain compression of neo-Hookean film bonded to an infinitely deep neo-Hookean substrate (no pre-stretch) (2012-2)

After the onset of period-doubling

$\mu_f / \mu_S = 1000, \varepsilon \approx 0.2$

$\mu_f / \mu_S = 15, \varepsilon \approx 0.2$

$\mu_f / \mu_S = 6, \varepsilon \approx 0.15$

For $\mu_f / \mu_S < 6$, a fold forms prior to period-doubling.

This is a form of localization
Period-Doubling:
Sun, Xia, Moon, Hwan & Kim (2012)

66 nm gold film on pre-stretched PDMS substrate

Period-Doubling:
Brau, Vandepaire, Sabbah, Poulard, Boudaoud & Damman (2011)

10-20 micron stiff film on PDMS
Role of Substrate Pre-stretch on Advanced Wrinkling Instabilities for Stiff Films on Compliant Substrates

Advanced Post-bifurcation Modes in Plane Strain Compression

Schematic of substrate pre-stretch used to produce compression in film when the substrate pre-stretch is released.

Un-stretched substrate  →  Stretched substrate  →  Film attached to stretched substrate  →  Stretch in substrate is relaxed to produce compression in film
Mountain ridges form (a new localization phenomenon!!)

Pre-stretch: $\lambda_0 = 2$

Ridge mode is observed for pre-stretches greater than 1.4 for both neo-Hookean and Arruda-Boyce elastomeric materials.
Experimental Observations of Localization in Pre-stretched film/substrate systems
Jianfeng Zang & Xuanhe Zhao (Duke U) (2012-3)

Typical sinusoidal mode with no localization

Localization, but not isolated ridges

Mode reverts to sinusoidal mode with further compression

Ebata, Croll & Crosby (2012) have observed ridges.
Why Folds not Ridges with pre-compression?

(2012-2)

\[ \Delta U^+ \]
\[ \Delta U^- \]
\[ \xi > 0 \]
\[ \xi < 0 \]
\[ |\xi| = 0.4 \]

For a pre-compression: \( \lambda_0 = 0.6 \)

\[ \frac{\Delta U^-}{\Delta U^+} = 0.92 \]

energy to form fold

energy to form ridge

For a pre-stretch: \( \lambda_0 = 2 \)

\[ \frac{\Delta U^-}{\Delta U^+} = 1.13 \]

energy to form fold

energy to form ridge

Why Ridges not Folds with pre-stretch?

Impose a ridge or fold displacement onto the surface of a pre-compressed or pre-stretched neo-Hookean substrate:

\[ u_1 = 0, \quad u_2 = 4\xi l \exp \left( -\left( \frac{x}{l} \right)^2 \right) \]

For pre-compression, it takes less energy to form a fold than a ridge

For pre-stretch, it takes less energy to form a ridge than a fold

These are nonlinear finite deformation effects inducing anisotropy in the substrate.
One slide on the role of pre-stretch on nonlinearity of neo-Hookean substrate (Hutch, unpublished)

An exact perturbation expansion (to second order) for sinusoidal surface tractions or displacements:

\[
\begin{align*}
U_1 &= \left( \ell / 2\pi \right) \left[ \xi \hat{u}_1^{(1)} \sin(2\pi x_1 / \ell) + \xi^2 \hat{u}_1^{(2)} \sin(4\pi x_1 / \ell) \right] \\
U_2 &= \left( \ell / 2\pi \right) \left[ \xi \hat{u}_2^{(1)} \cos(2\pi x_1 / \ell) + \xi^2 \left( \hat{u}_2^{(2)} \cos(4\pi x_1 / \ell) - \hat{u}_1^{(1)} \hat{u}_2^{(1)} / 2 \right) \right] \\
T_1 &= \left( \mu_s / \lambda_3 \right) \left[ \xi \hat{t}_1^{(1)} \sin(2\pi x_1 / \ell) + \xi^2 \hat{t}_1^{(2)} \sin(4\pi x_1 / \ell) \right] \\
T_2 &= \left( \mu_s / \lambda_3 \right) \left[ \xi \hat{t}_2^{(1)} \cos(2\pi x_1 / \ell) + \xi^2 \hat{t}_2^{(2)} \cos(4\pi x_1 / \ell) \right]
\end{align*}
\]

\[
\hat{i}_\alpha^{(1)} = C_{\alpha\beta}(r) \hat{u}_\beta^{(1)} , \quad \hat{i}_\alpha^{(2)} = 2C_{\alpha\beta}(r) \hat{u}_\beta^{(2)} - C_{\alpha\beta\gamma}(r) \hat{u}_\beta^{(1)} \hat{u}_\gamma^{(1)}
\]

Pre-stretch ratio: 
\[
r = \frac{\lambda_2}{\lambda_1} = \frac{1}{\lambda_1^2 \lambda_3}
\]

For: 
\[
\bar{T}_2(x_1) = \bar{T}_2(0) \cos(2\pi x_1 / \ell)
\]

\[
\frac{U_2(0)}{\ell} = \left( \frac{\bar{T}(0)}{2\pi \mu_s / (D_{22} \lambda_3)} \right) - K \left( \frac{\bar{T}(0)}{2\pi \mu_s / (D_{22} \lambda_3)} \right)^2
\]

\[
T_1 = 0
\]

\[
U_1 = 0
\]

\[
T_1 = 0
\]

\[
U_2(0) / \ell
\]
Conclusions, Analogies and Speculations

• The only buckling problems with comparable instability and imperfection-sensitivity (known to me) are the cylindrical shell under axial compression and the spherical shell under external pressure. Both have many modes associated with the critical stress. For both of these problems, the critical modes are never observed experimentally (except possibly by high speed camera).

• The wrinkling mode is so unstable and so imperfection-sensitive that it is seems likely imperfections will always be present leading to dynamic wrinkling instability and evolving into a crease at strains below the Biot wrinkling strain. It is highly unlikely that the wrinkling mode can be observed except possibly if captured by a high speed camera.

• Wrinkling and creasing are different from cylindrical and sphere shell buckling in one very important respect—They can have any length scale. In principle, a perfectly flat surface should reach the Biot wrinkling strain before becoming unstable, but in practice is seems reasonable to expect that imperfections will always be present at some scale to trigger creases at the creasing strain.

• An open question: What is the length scale at which some material effect (e.g., strain gradient effects, surface oxidation, etc.) comes into play to set a minimum size for wrinkles and creases?
  
  See PRL paper by Chen, Cai, Suo & Hayward (2012) for one answer (surface tension).

There is more!
Other Systems with Buckling Phenomena In Common with Wrinkling Instabilities

Spherical shell buckling arrested by internal mandrel: Without mandrel this mode would not be observed—Like non-observability of wrinkles on a homogeneous substrate.

Experimental buckling loads of elastic cylindrical shells under axial compression normalized by bifurcation load of perfect shell—Like wrinkling of homogeneous substrate, bifurcation strain is effectively unreachable.

Railroad track buckling due to compression induced by heating in the sun—Note the localization, analogous to ridge formation in thin film wrinkling.
An example from M.-Y. Moon of wrinkling on a prolate spheroid substrate (Diamond-like carbon (DLC) on PDMS)

DLC on Spherical geometry
But thickness of DLC might be not uniformed on the surface of PDMS sphere

\[ t_{DLC} \approx 10 \text{nm}, \quad R_{PDMS} \approx 50 \mu\text{m} \]

wavelength \( \sim 2 \mu\text{m} \)
Observations of various patterns of a silica-like film on a PDMS substrate formed by ultraviolet-ozone oxidation. Compressive stress induced by mismatch in film/substrate swelling due to ethanol vapor absorption.

Umass system (Breid & Crosby) (2011-5)

1D mode
plane strain compression

Square checkerboard mode
(different system)
Equi-biaxial compression

Hexagonal mode
Equi-biaxial compression

Herringbone mode
Equi-biaxial compression

Films thickness in range from 100 to 200 nm.
Critical periodic modes plus the herringbone mode (2011-5)

\[ \sigma_c = \frac{\bar{E}_f}{3} \left( \frac{E_s^*}{\bar{E}_f} \right)^{2/3} / 4 \]

All modes satisfying: \( w = \frac{\sin(k_1 x_1)}{\cos(k_1 x_1)} \times \frac{\sin(k_2 x_2)}{\cos(k_2 x_2)} \) with \( \sqrt{k_1^2 + k_2^2} = k = t^{-1} \left( \frac{3 E_{s}^*}{\bar{E}} \right)^{1/3} \)

1D: \( w = \cos(kx_1) \)

Square checkerboard:
\( w = \cos(kx_1 / \sqrt{2}) \cos(kx_2 / \sqrt{2}) \)

Hexagonal:
\( w = \cos(kx_1) + 2 \cos(kx_1 / 2) \cos(\sqrt{3}kx_2 / 2) \)

Triangular:
\( w = -\sin(kx_1) + 2 \sin(kx_1 / 2) \cos(\sqrt{3}kx_2 / 2) \)

Herringbone (not critical):
\( w = \xi_1 \cos(\beta kx_1) + \xi_2 \sin(\beta kx_1) \cos(\beta kx_2) \)
Energy in buckled state for all the modes (linear substrate) (2011-5)
Mode transitions under increasing over stress: Observed & Predicted (2011-5)

\[ \sigma_0 \sim \text{equi-biaxial stress in unbuckled film} \]
\[ \sigma_C = \bar{E}_f \left(3 \bar{E}_s / \bar{E}_f \right)^{2/3} / 4 \sim \text{Critical equi-biaxial stress at onset of buckling} \]
\[ \bar{E} = E / (1 - \nu^2) \]

In region of low over stress, the hexagonal mode is always observed in the UMass system.

**Predicted** transitions based on min. energy for flat systems: unbuckled, square, herringbone

**Observed** transitions for UMass system:: unbuckled, hexagonal, herringbone

Transition from hexagonal pattern to herringbone pattern by coalescence of neighboring hexagonal cells