Thin Film Herringbone Buckling Patterns

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ABSTRACT
Thin metal film deposited on compliant substrate undergoes equi-biaxial compression and buckles into a highly ordered herringbone pattern \([1,2]\). In this study, it is shown that compare with the bifurcation competing modes, the herringbone mode has the lowest strain energy and therefore it is the preferred buckling pattern in the thin film.

INTRODUCTION
Self-assembly involving thin films has important applications in microfabrication. Recent studies \([1,2]\) have shown that simply by elastic buckling, thin film on compliant substrate can form highly ordered patterns with distinctive features. Compare with the traditional complicated physical or chemical processes, the mechanical buckling is perhaps the cheapest and quickest way to form an ordered structure spontaneously in the thin film.

Due to the large thermal expansion mismatch, thin metal film vapor deposited on a thick compliant elastomer substrate undergoes equi-biaxial compression when the system is cooled. The film buckles into periodic structures with distinctive features when the temperature variance exceeds a critical number. The buckling patterns can be manipulated by creating non-planar substrate topography prior to deposition \([1,2]\). In this case, the self-assembled structure in thin film conforms to the underlying substrate profile. The most intriguing phenomenon happens when the substrate is not pre-patterned. As shown in Figure 1, for the system where a 50nm Au film is vapor deposited on a flat thick PDMS substrate, the highly ordered herringbone mode arises when the temperature is lowered \([1,2]\). Here, The crest-to-crest separation, or the buckle wavelength is about 30\(\mu\)m, and the distance between jogs in the herringbone mode is about 100\(\mu\)m. The change in direction of the waves at each jog is approximately 90\(^\circ\). The amplitude of the waves is about one micron when the temperature variance is 100\(^\circ\)C. Thus, although the amplitude is large compared to the film thickness, the mode is shallow in the sense that the slopes of the pattern are small. The herringbone buckle pattern diminishes when the temperature is elevated to the deposition temperature and the film remains attached with the substrate. This indicates that the buckling problem formulated here is elastic with no delamination. The strains associated with the buckling mode are also small, and both the film and the substrate materials are within their respective linear elastic ranges.

When the substrate is initially flat, the herringbone buckle pattern appears to be the preferred buckling mode when the system has been cooled well below the onset of buckling. It is very different from any mode obtained from the classical linear bifurcation analysis, which is due to the highly nonlinear nature of the system as the temperature drops well below critical. Compare with other competing bifurcation modes, such as the one-dimensional mode and the checkerboard mode sketched in Figure 1, the herringbone mode has the ability to reduce the overall in-plane stress in the film in all directions and its near-inextensionality induces only very little stretching energy. As will be shown later, these two characteristics ensure the herringbone
mode to be the minimum energy configuration among its competitors (c.f. Figure 1), which offers the clue for its existence.

LINEARIZED STABILITY ANALYSIS OF THE CHECKERBOARD MODE

In this section, the attention is confined to the critical buckling stress associated with the periodic checkerboard mode. Denote the Young’s modulus, Poisson’s ratio and coefficient of thermal expansion of the film are by \( E \), \( \nu \), and \( \alpha \), respectively. The corresponding substrate properties are \( E_s \), \( \nu_s \) and \( \alpha_s \). All numerical results in this study are presented for Au film deposited on PDMS substrate, where \( \nu = 1/3 \), \( \nu_s = 0.48 \), \( \alpha_s / \alpha = 1/20 \) and \( E_s / E = 4100 \). Solutions for other film/substrate systems can easily be obtained by similar approaches. The film thickness is \( t \).

The substrate is assumed to be infinitely thick who imposes its in-plane strains on the film. The film is represented as an infinite elastic thin plate subject to equi-biaxial compression \( \sigma_0 \) satisfying the linearized von Karman plate equation [3]:

\[
D \nabla^4 w + \sigma_0 \nabla^2 w = -p
\]

(1)

Here, \( \nabla^4 \) is the bi-harmonic operator, \( D = E t^3 /[12(1 - \nu^2)] \) is the bending stiffness of the plate, \( w \) is its displacement perpendicular to the plane, \((x_1, x_2)\), \( p \) is the stress component acting perpendicular to the plate that is exerted by the deformed substrate. Assuming the temperature of the system is reduced by \( \Delta T \), if the film is elastic and unbuckled, the equi-biaxial pre-stress \( \sigma_0 \) is then given by

\[
\sigma_0 = [E(1 - \nu)]\Delta \alpha \Delta T
\]

(2)

where \( \Delta \alpha = \alpha_s - \alpha \). By ignoring the tangential traction exerted by the substrate, eq. (1) admits periodic checkerboard solutions of the form

\[
w = \hat{w} \cos(k_1 x_1) \cos(k_2 x_2), \quad p = \hat{p} \cos(k_1 x_1) \cos(k_2 x_2)
\]

(3)

where from 3D elasticity \( \hat{w} = 2 \hat{p} / (\bar{E}_s k) \) with \( k = \sqrt{k_1^2 + k_2^2} \) and \( \bar{E}_s = E_s / (1 - \nu_s^2) \). Substituting (3) into (1) leads to the eigenvalue equation \( \sigma_0 j = D k^2 + \bar{E}_s / 2 k \). Finally, the critical buckling stress

\[
\sigma_0^c = \left( \frac{3 \bar{E}_s}{E} \right)^{2/3}
\]

is obtained at the critical buckling wave number \( k^c t = (3\bar{E}_s / E)^{1/3} \), where \( E = E/(1 - \nu^2) \). This is the result for the one-dimensional, plane strain wrinkling stress, which is widely known [4].

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**Figure 1.** The herringbone buckling pattern (on the left) versus the competing one-dimensional and checkerboard buckling modes (on the right).
The compressed film in the equibiaxial state has multiple bifurcation modes satisfying
\[ \sqrt{k_1^2 + k_2^2} = k^c t = (\frac{3E_i}{E})^{1/3} \]. We limit our attentions to both the one-dimensional mode \((k_1 = k^c, k_2 = 0)\) and the square checkerboard mode \((k_1 = k_2 = k^c / \sqrt{2})\). They are the competing modes of the herringbone mode.

NONLINEAR POST-BUCKLING ANALYSIS OF THE ONE-DIMENSIONAL MODE

When \(k_2 = 0\), the checkerboard mode (3) reduces to the one-dimensional mode, such that the normal displacement in the finite amplitude state is now
\[ w = \hat{w} \cos(k_1 x_1) \]
where \(k_1\) is an arbitrary nonzero wave number. Compatibility dictates that

\[ \frac{1}{E_t} (\sigma_{t} - \sigma_0^E) t = \frac{k_1}{4\pi} \int_0^{\frac{2\pi}{k_1}} \left( \frac{d\hat{w}}{dx_1} \right)^2 \, dx_1 \]

where \(\sigma_0^E = Dk_1^2 / t + E_i / (2k_1 t)\) is the stress at the onset of buckling associated with \(k_1\). From (4), the amplitude of the one-dimensional buckling mode is derived:

\[ \frac{\hat{w}}{t} = \frac{2}{k_1} \sqrt{\frac{(\sigma_0 - \sigma_0^E)}{E}} \]

Results will be presented for various \(k_1\) including the critical case with \(k_1 = k^c\) and \(\sigma_0^E = \sigma_0^{C^c}\). The solution is produced for temperatures such that \(\sigma_0 > \sigma_0^E\). If the film is unbuckled, the energy per unit area in the film/substrate system is \(U_0 = (1 - \nu) \sigma_0^E \bar{t} / E\). In the buckled state, the elastic strain energy of the system \(U\) has contributions from the stretching energy of the film, the bending energy of the film, and elastic energy stored in the substrate. In the same order of these three contributions, the energy ratio between the buckled and unbuckled states is:

\[ \frac{U}{U_0} = \frac{1 + \nu}{2} \left( \frac{\sigma_0^E}{\sigma_0} \right)^2 + (1 - \nu)^2 \left( \frac{E_i}{E} \right)^2 \left( \frac{\hat{w}}{t} \right)^2 + \frac{(1 + \nu)k_1 t}{8} \left( \frac{E_i}{E} \right)^2 \left( \frac{\hat{w}}{t} \right)^2 \]

Plots of \(U / U_0\) as a function of \(\sigma_0 / \sigma_0^{C}\) are given in Fig. 2 for a gold film on a PDMS substrate [1]. Results for the normalized energy are presented for five values of the wavelength ratio, \(L / L^c = k^c / k_1\), where \(L = 2\pi / k_1\) is the wavelength and \(L^c = 2\pi / k^c\) is the wavelength of the critical mode. The results of Fig. 2 for \(U / U_0\) for the one-dimensional mode show that the lowest energy state is associated with the critical mode \((L / L^c = 1)\) even at finite amplitude buckling deflections. Thus, the results of Fig. 2 emphasize the strong preference for the wavelength associated with the critical wavelength \(L^c = 2\pi / k^c\) when the mode is one-dimensional.
NUMERICAL ANALYSIS OF THE SQUARE CHECKERBOARD AND HERRINGBONE MODES

The finite element method is used to study the post-buckling behavior of the square checkerboard and herringbone modes. The calculation is carried out by using the commercial code ABAQUS [5]. Within the three-dimensional periodic cell, the film is represented by 1000 three-dimensional eight-node, quadratic thin shell elements that account for finite rotations of the middle surface. The semi-infinite substrate is meshed with 20-node quadratic block elements with reduced integration. The film/substrate system is assigned a thermal expansion mismatch, \( \Delta \alpha \), and a temperature drop \( \Delta T \) is imposed starting from the unstressed state. The biaxial compressive stress in the unbuckled film is given by (2).

For each mode, a unit periodic cell is identified and meshed with periodicity conditions imposed on the edges of cell, both for the film and the underlying substrate. The unit cell of the square checkerboard is a semi-infinite parallelepiped with cross-section \( L \times L \). The unit top surface of the periodic cell for the herringbone mode is shown in Fig. 3. The parallelepiped is characterized by its width (half-jog spacing), \( a \), breadth (wavelength), \( L \), and inclination angle (jog angle), \( \alpha \). In both modes, a very small initial imperfection is prescribed such that the plate at \( \Delta T = 0 \) has a slight middle surface deflection of 2% of the film thickness.

Contour plot of the normal deflection of the film \( w \) within the cell of the herringbone mode is displayed in Fig. 4 for \( L / L^c = 1 \) and \( \sigma_0 / \sigma_0^c = 26 \), with the cell dimensions \( a / L = 2 \) and \( \alpha = 45^\circ \). It will be seen below that the minimum energy configuration corresponds to \( L / L^c \approx 1 \), and for this value it can be seen that the deflection shape displays the features of the herringbone mode seen in Fig. 1. The mode has a curving ridge running along the center of the cell that aligns itself to merge smoothly at the jog with the ridge in the next cell.

THE ENERGY COMPETITION

We address two outstanding problems in this section: 1) Does the herringbone mode have the minimum energy configuration among the competing modes? 2) What is the optimum geometry of the herringbone pattern whose strain energy is minimum?

When the wavelength is set to be equal to the critical wavelength at the onset of buckling, the computed average strain energies per area in the film/substrate system are presented in Fig. 5 in normalized form as \( U / U_0 \) versus \( \sigma_0 / \sigma_0^c \). It can be readily seen that among the three buckling modes considered, the herringbone mode produces the lowest average elastic energy of the film/substrate system for films stressed well above critical. The

\[
\begin{align*}
\alpha &= 45^\circ, \quad a / L = 2 \\
\sigma_0 / \sigma_0^c &= 26 \\
&\begin{array}{c|c}
L / L^c & w / t \\
\hline
1 & -4.2 \\
2 & -2.5 \\
3 & -0.8 \\
4 & 0.8 \\
5 & 2.5 \\
6 & 4.2 \\
\end{array}
\end{align*}
\]
herringbone mode is able to relax the biaxial pre-stress stress, \( \sigma_0 \), in the film in all directions while inducing relatively little concurrent stretch energy localized in the jog regions. It is shown from Fig. 5 that these unique characteristics have made the herringbone mode to become the preferred buckling pattern in terms of minimum energy. By contrast, the one-dimensional mode requires essentially no stretch energy (it continues to exhibit zero Gaussian curvature), but it relaxes the biaxial pre-stress only in one direction. The checkerboard mode relaxes the pre-stress in all directions, but it develops non-zero Gaussian curvature (c.f. Fig. 1) and induces much more concurrent stretch energy than the herringbone mode.

The dependence of \( U / U_0 \) on the parameters of the cell geometry \( (L/L^C, a/L, \alpha) \) is presented in Fig. 6. Fig. 6a shows that the minimum energy is associated with \( L/L^C \sim 1 \). The energy of modes with \( L/L^C > 1 \) or \( L/L^C < 1 \) is distinctly above the minimum. In Fig. 6b it is seen that the energy in the buckled state weakly dependent on the jog spacing, \( a/L \). Only for very short cells, \( a/L = 0.5 \), is the energy noticeably above the minimum. Evidence for this insensitivity is reflected in the experimental herringbone patterns in Fig. 1, where it can be seen that the distance between jogs varies by at least a factor of three from one section of the film to another. Similarly, there is not a very strong dependence of the energy of the buckled system on the inclination of the cell, \( \alpha \), although the minimum is attained for \( \alpha \approx 45^\circ \) (Fig. 6c), which is in agreement with experimental evidence shown in Fig. 1.

A feature seen in both Figs. 5 and 6 is that the ordering of the relative energy trends is invariant of \( \sigma_0 / \sigma_0^C \). Thus, the parameters governing the geometry of the minimum energy mode do not change in a significant way as \( \sigma_0 / \sigma_0^C \) (or \( \Delta T \)) increases. It is worthy to notice that the herringbone mode is not a bifurcation mode, and it only becomes a preferred mode in the sense of having minimum energy at \( \sigma_0 / \sigma_0^C \) somewhat above unity. At \( \sigma_0 / \sigma_0^C = 1 \), the amplitude and normalized energy of the herringbone mode is dominated by the initial imperfection.

**CONCLUSIONS**

In this study, we have shown that the herringbone mode consists the lowest strain energy among several competing modes. Therefore, herringbone is the preferred buckling mode when the equi-biaxial compressive stress is above the critical level. The minimum energy state of the herringbone mode has undulation wavelength very close to the critical wavelength of the one-dimensional mode, \( L^C = 2\pi (\bar{E}/3\bar{E}_s)^{1/3} \), and jog angle \( \alpha \approx 45^\circ \). The minimum energy state is
weakly dependent on the jog spacing $a$. These features predicted by the theoretical minimum energy state are in reasonable agreement with the experimental herringbone patterns [1,2].

References