Electronic Supplementary Material (ESI) for Soft Matter

Supplemental Information 1: Wrinkling Wavelength

Wrinkling wavelength is dictated by film thickness t and the modulus of both film (E_f) and the substrate (E_s)[1].

$$\lambda \sim 2\pi t \left(\frac{E_f}{3E_s}\right)^{1/3} \tag{S1}$$

To demonstrate that the wrinkles observed in this experiment follow the classical wrinkling behavior described in equation (S1), the wavelength of wrinkles for varying film thickness for the substrates with elastic modulus of 1.3 MPa (left) and 0.7 MPa (right) are shown in the figure below. The wrinkling wavelength was measured at the onset using optical profilometer or optical microscope with Fast Fourier Transform. The plot of wavelength with respect to thickness shows a linear trend, which follow the scaling of equation (S1), and the slope of the linear fit equals $2\pi(\frac{E_f}{3E_s})$.

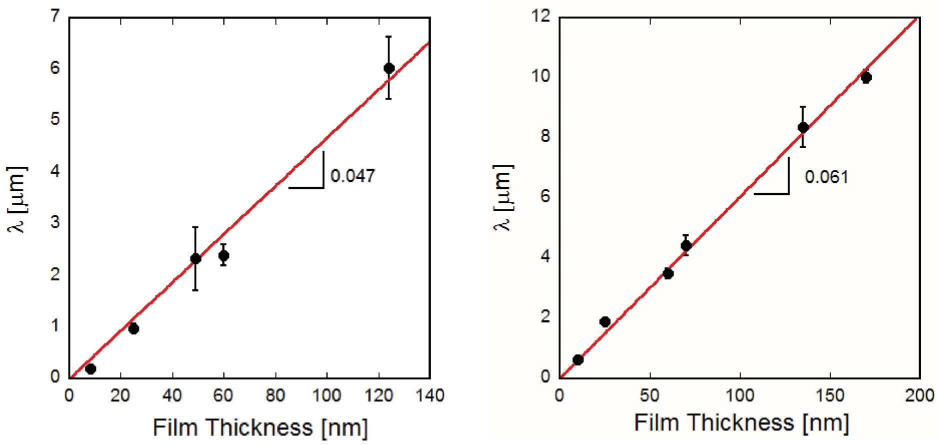

FIG. S1: Wrinkling wavelength for PDMS substrates with varying polystyrene film thickness. The black circle markers are the average values of wrinkling wavelength for each film thickness and the red line is the linear fit to our experimental data. The modulus of the PDMS substrate is varied from 1.3 MPa (left) to 0.7 MPa (right).

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Supplemental Information 2: Atomic Force Microscope Image

The cross-section data shown in figure 3 was obtained using optical profilometer. To confirm the shape of wrinkles and folds, we also observed using atomic force microscope. For this sample, 45 nm thick film was placed on 20:1 PDMS. The applied global strain was 0.04, which is past the average fold critical strain of 0.03.

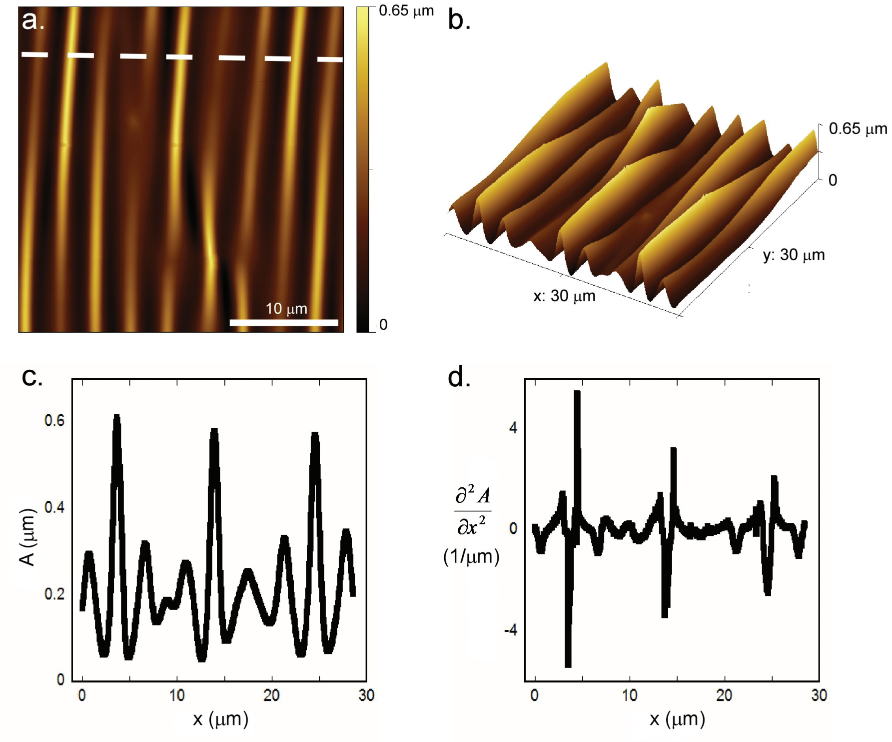

FIG. S2: (a) and (b) show the two-dimensional and three-dimensional AFM images, respectively. Cross-section amplitude data shown in (c) is obtained at the dotted line in (a). (d) is the plot of second derivative of amplitude with respect to the distance.

As shown in figure S2c, the amplitude of fold is larger than wrinkles next to it, and figure S2d shows that there is a large negative curvature at the peak of the fold. Figure S2d was obtained by taking numerical derivatives of the cross-section data.

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Supplemental Information 3: Code for Amplitude Histogram Plot

The amplitude data was taken using optical profilometer, and Matlab®program was written to construct amplitude histogram plot from the three-dimensional data. An example of the Matlab code used for horizontally aligned buckled structures is shown below.

```
%This function takes in 3D data (x,y,z) and finds local maximum and minimum.
%After that, it finds the amplitudes and plots them in histogram format.
%only works when the wrinkles are formed horizontally.
z = sample1_1; %file name
x = [1:1:640]; %number of points on x
y = [1:1:480]; %number of points on y
A = zeros(480,640);
for j = 1:1:640;
    z_j = z(:,j);
    N = length(y)-2;
    h = y(2)-y(1);
    f = zeros(N+2,1);
    f(1) = (1./(2.*h)).*((-3.*z_j(1)))+(4.*z_j(2))-(z_j(3));
    f(2) = (1./(2.*h)).*((-3.*z_j(2)))+(4.*z_j(3))-(z_j(4));
    f(N+1) = (1/(2.*h)).*(z_j(N-1))-4.*z_j(N)+3.*z_j(N+1);
    f(N+2) = (1/(2.*h)).*(z_j(N))-4.*z_j(N+1)+3.*z_j(N+2);
    for k=3:N
    f(k)=(1./(12.*h)).*(z_j(k-2)-8.*z_j(k-1)+8.*z_j(k+1)-z_j(k+2));
    %First derivative approximation
    end
    index_j = find(f(1:end-1).*f(2:end)<0);
    %Find where the first derivative crosses zero
    d_j = z_j(index_j);
    %Find the index number where the first derivative crosses zero
    for i = 1:length(d_j)-2
```

```
        A(index_j(i),j) = (abs(d_j(i+1)-d_j(i))+abs(d_j(i+2)-d_j(i+1)))/2;
        %amplitude values between local max and min
    end
end
t = nonzeros(A);
s = mode(mode(t));
[u,v]=hist(t,10000);
g = max(u);
h = (u./g);
bar(v,h,0.1)%plotting histogram
```

Supplemental Information 4: Critical Strain Equation for Delamination

The critical strain equation for delamination is derived by following the energy approach by Vella, et al.[2]. For a stiff thin film with thickness t placed on a compliant substrate with thickness h, the total elastic energy U for n identical blisters with height δ and width λ is a combination of the energy contribution due to bending of the thin film and the elastic energy of the substrate that is localized directly underneath the blisters:

$$U = n\left(\pi^4 B\frac{\delta^2}{\lambda^3}w + \alpha E_s \epsilon^2 \lambda^2\right) \tag{S2}$$

where B is the bending stiffness of the film, w is the width of the film, and E_s is the substrate elastic modulus. In our experimental system, the blisters width is on the order of m, whereas the width of the film as well as the substrate thickness h is on the order of mm. We consider the small blisters regime where $\lambda \ll w, h$. Using the blister profile, geometrical relationship is given by:

$$\frac{\Delta L}{n} = \frac{\pi^4}{4}\frac{\delta^2}{\lambda} \tag{S3}$$

By minimizing the elastic energy with respect to n, critical value of $\Delta L/n$ is:

$$\frac{\Delta L_c}{n} = \left(\frac{B\Delta\gamma^2 w^3}{\alpha^3 E_s^3}\right)^{1/5} \tag{S4}$$

If $L_0 \sim n\lambda$ is assumed:

$$\epsilon_d \sim \frac{\Delta L_c}{L_0} \sim \frac{1}{\lambda_c}\left(\frac{B\Delta\gamma^2 w^3}{\alpha^3 E_s^3}\right)^{1/5} \tag{S5}$$

For small blisters, $\lambda_c \sim \left(\frac{B^2}{E_s \Delta\gamma}\right)^{1/5}$ and $\alpha \sim w$[2]. Since $\Delta\gamma \sim G_c$ and $B \sim E_f t^3$, equation (S5) is rewritten as:

$$\epsilon_d \sim \left(\frac{G_c}{E_s t}\right)^{3/5}\left(\frac{E_s}{E_f}\right)^{1/5} \tag{S6}$$

Supplemental Information 5: Fast Fourier Transform (FFT) of the optical images

Fast Fourier Transform of a representative sample is presented here. 25 nm thick film is placed on 10:1 PDMS and strain was applied up to 0.07.

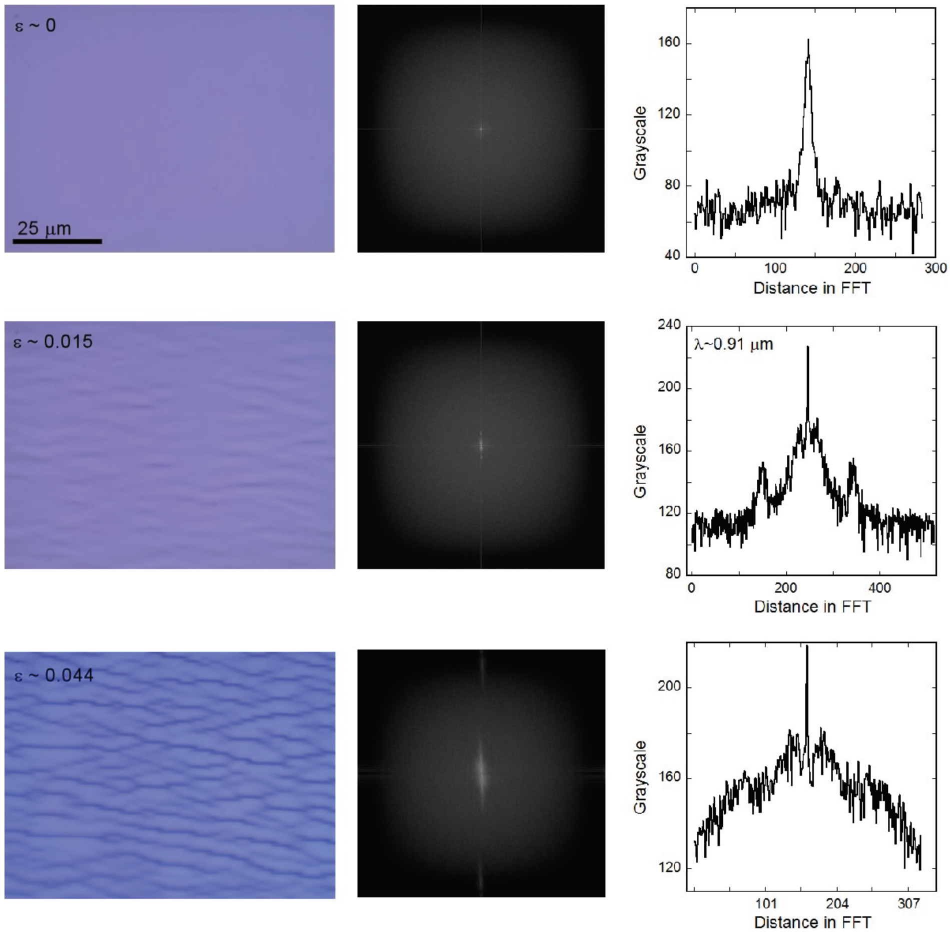

FIG. S3: Optical images and FFT images of a representative sample with 25 nm thick film placed on 10:1 PDMS. Strain was applied to this sample up to 0.07. FFT was performed using Image Processing and Analysis in Java(ImageJ).

There is no structure observed with no applied strain. When applied strain was 0.015, wrinkles start to form across the surface and peaks are observed on the FFT image. As folds started to emerge at an applied strain of 0.044, there is a broadening of the peaks.

[1] J. Genzer and J. Groenewold, Soft Matter **2**, 310 (2006).

[2] D. Vella, J. Bico, A. Boudaoud, B. Roman, and P. M. Reis, Proceedings of the National Academy of Science of the United States of America **106**, 10901 (2009).