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Pre-tension regulates buckling patterns of soft films with interactions

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Abstract – We study elastic buckling of a uni-axially pre-tensioned soft elastic film in proximity to a rigid contactor due to van der Waals interaction. By a linear analysis, we show that the film will buckle into stripes parallel to the tensile direction, with tunable spacing governed by the magnitude of the pre-tension. Such a regulating effect of pre-tension may find practical applications in generating precise surface patterns.

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Surface buckling of soft elastic solids has been an important topic of research [1,2]. The study is not only of fundamental interest for understanding the mechanisms themselves, but also of technological importance for generating precise patterns in practical applications [3–5]. Recently, experimental observation [6] indicated that, when brought in proximity to a plane contactor, the surface of a soft thin elastic film bonded to a rigid substrate will lose planarity and jumps in adhesion to the contactor in a periodic way. The ripple pattern exhibits fixed length scale of the same order as the film thickness, but lacks a long-range order. Theoretically, this phenomenon has been interpreted to be a result of the interplay of the elastic energy of the thin film and the interaction energy between the film and the contactor [7–15]. The interaction may arise from any of such causes as the long-range van der Waals force and external electric field, etc. A question is raised naturally: can we regulate the buckling pattern in a simple way? In this letter, we show that the buckling pattern can be regulated indeed by pre-tensioning the thin film.

Our idea can be explained qualitatively. Suppose that an elastically isotropic and incompressible film of thickness H is tensioned in one direction up to λ times its original length, and then glued perfectly onto a flat, rigid substrate. The dimensionless parameter λ, called principal stretch in the tensile direction, characters the deformation of the homogeneously tensioned film. Due to the confinement of the substrate, the tensioned state of the film can be retained after removing the external force. As a consequence of incompressibility, the film possesses pre-stretch $\lambda^{-1/2}$ in the plane normal to the tensile direction and, especially, its thickness now becomes $h = \lambda^{-1/2}H$. Intuitively, a further stretch of the film surface normal to the pre-tension direction is easier than parallel to the pre-tension direction. We thus expect that the pre-tension can regulate the buckling mode of the film when a flat contactor is brought in enough proximity.

To perform a quantitative analysis, we start with the consideration of the homogeneously tensioned state of the film. In the absence of a nearby contactor, the film surface is planar. We choose this state of the film as the reference configuration, and introduce a rectangular coordinate system (x_1, x_2, x_3) so that the (x_1, x_2)-plane coincides with the film surface and the x_1-axis is along the tensile direction (fig. 1). When a flat contactor is fixed at a distance d above the film, an additional displacement $\mathbf{u} = u_i \mathbf{e}_i$ is induced due to van der Waals interaction, with $\mathbf{e}_i$ being the base vector along the x_i-axis. We assume that $\mathbf{u}$ is sufficiently small for the sake of a linear analysis, but the principal stretch λ can be significantly greater than unity. Thus, the film undergoes a finite pre-tension followed by a small displacement perturbation. This is in strong contrast to the previous studies [7–15] in which the film does not bear any pre-tension.

For purpose of deriving the basic equations of the film, we denote the position vectors of a material point in the stress-free state, the homogeneously tensioned

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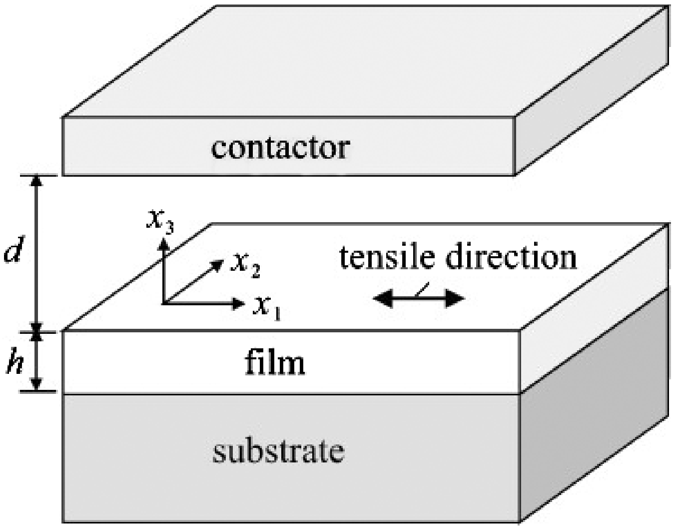

Fig. 1: Sketch of the problem, where the elastomer film is pretensioned in the x_1-direction.

state, and the current state by $\mathbf{X} = X_i \mathbf{e}_i$, $\mathbf{x} = x_i \mathbf{e}_i$, and $\hat{\mathbf{x}} = \hat{x}_i \mathbf{e}_i$, respectively. Since $x_1 = \lambda X_1$, $x_2 = \lambda^{-1/2} X_2$, $x_3 = \lambda^{-1/2} X_3$, and $\hat{x}_i = x_i + u_i$, $\mathbf{X}$ and $\hat{\mathbf{x}}$ can be represented as functions of $\mathbf{x}$, and the total deformation gradient of the film can be obtained by $\mathbf{F} = \partial \hat{\mathbf{x}} / \partial \mathbf{X} = \mathbf{F}_2 \cdot \mathbf{F}_1$, in which

$$\begin{aligned} &\mathbf{F}_1 = \partial \mathbf{x} / \partial \mathbf{X} = \lambda \mathbf{e}_1 \otimes \mathbf{e}_1 + \lambda^{-1/2} \mathbf{e}_2 \otimes \mathbf{e}_2 + \lambda^{-1/2} \mathbf{e}_3 \otimes \mathbf{e}_3, \\ &\mathbf{F}_2 = \partial \hat{\mathbf{x}} / \partial \mathbf{x} = \mathsf{I} + \nabla \mathbf{u}. \end{aligned} \tag{1}$$

In the above relations, the symbol "$\otimes$" stands for tensor product, I is the identity tensor, $\nabla \mathbf{u} = u_{i,j}\, \mathbf{e}_i \otimes \mathbf{e}_j$ is the displacement gradient, and a comma is used to denote differentiation with respect to the suffix coordinate. Of special importance are the left and right Cauchy-Green strain tensors, defined by $\mathbf{B} = \mathbf{F} \cdot \mathbf{F}^T$ and $\mathbf{C} = \mathbf{F}^T \cdot \mathbf{F}$, respectively, with T representing the transpose of a tensor. Either $\mathbf{B}$ or $\mathbf{C}$ can completely describe the finite strain of the film. For simplicity, we assume that the film material is neo-Hookean [16]. In this situation the constitutive law of the film is written as $\boldsymbol{\sigma} = \mu \mathbf{B} - p\, \mathsf{I}$, where $\boldsymbol{\sigma}$ is the Cauchy stress tensor defined with respect to unity area in the current state of the film, μ is shear modulus, and p is hydrostatic pressure. Making use of eq. (1) and keeping only the first-order term of the displacement gradient, we have

$$\boldsymbol{\sigma} = \mu \left[\mathbf{B}_1 + \mathbf{B}_1 \cdot (\nabla \mathbf{u})^T + (\nabla \mathbf{u}) \cdot \mathbf{B}_1 \right] - p\, \mathsf{I}, \tag{2}$$

in which $\mathbf{B}_1 = \left(\lambda^2 - \lambda^{-1}\right) \mathbf{e}_1 \otimes \mathbf{e}_1 + \lambda^{-1} \mathsf{I}$. When $\lambda \neq 1$, we can see the stress $\boldsymbol{\sigma}$ as a function of the tensor $\nabla \mathbf{u}$ and the scalar p as given in (2) does not satisfy $\mathbf{Q} \cdot \boldsymbol{\sigma}(\nabla \mathbf{u}, p) \cdot \mathbf{Q}^T = \boldsymbol{\sigma}(\mathbf{Q} \cdot \nabla \mathbf{u} \cdot \mathbf{Q}^T, p)$ if the orthogonal tensor $\mathbf{Q}$ is chosen as $\mathbf{Q} = \mathbf{e}_1 \otimes \mathbf{e}_2 - \mathbf{e}_2 \otimes \mathbf{e}_1 + \mathbf{e}_3 \otimes \mathbf{e}_3$. This implies that the pre-tension leads to that the originally isotropic film material behaves anisotropically with respect to the reference configuration. Note that the neo-Hookean constitutive law (2) is an approximation valid for principal stretches close to 1. However, we believe that we can use it to show an informative trace of the real physics, though some more elaborated constitutive laws have been proposed for soft elastic materials.

The free energy Π of the system involves the contributions from the strain energy, the surface energy, and the van der Waals interaction. Omitting the effect of body force and denoting volume and surface elements in the reference configuration by dv and ds, respectively, we have

$$\begin{aligned} \Pi = &\int_v \Sigma(\mathbf{E}) \det(\mathbf{F}_2) \mathrm{d}v \\ &+ \int_s [\gamma + U(\mathbf{u} \cdot \mathbf{e}_3)] \det(\mathbf{F}_2)(\mathbf{e}_3 \cdot \mathbf{C}_2^{-1} \cdot \mathbf{e}_3)^{1/2} \mathrm{d}s. \end{aligned} \tag{3}$$

Here $\Sigma(\mathbf{E})$ is the strain energy density depending on the Lagrangian strain tensor $\mathbf{E}$ measured with respect to the stress-free state, γ is the specific surface energy that is assumed constant, $U(\mathbf{u} \cdot \mathbf{e}_3) = -A/[12\pi(d - \mathbf{u} \cdot \mathbf{e}_3)]^2$ is the van der Waals interaction potential with A being the Hamaker constant, and $\mathbf{C}_2 = \mathbf{F}_2^T \cdot \mathbf{F}_2$ is the additional right Cauchy-Green strain tensor caused by the van der Waals interaction. The Lagrangian strain tensor $\mathbf{E}$ relates to the total right Cauchy-Green strain tensor by $\mathbf{E} = (\mathbf{C} - \mathsf{I})/2$, and the variation of strain energy density $\Sigma(\mathbf{E})$ can be expressed by $\delta\Sigma(\mathbf{E}) = \mathbf{T} : \delta\mathbf{E}$, in which $\mathbf{T} = \det(\mathbf{F}) \mathbf{F}^{-1} \cdot \boldsymbol{\sigma} \cdot \mathbf{F}^{-T}$ is the second Piola-Kirchhoff stress tensor. Recalling that the film material is incompressible and the displacement is small, we can infer from eq. (1) that $\det(\mathbf{F}_2) = 1$, $\det(\mathbf{F}) = 1$, $\mathbf{F}_2^{-T} = \mathsf{I} - (\nabla \mathbf{u})^T$ and $\mathbf{T} : \delta\mathbf{E} = \boldsymbol{\sigma} : \delta(\nabla \mathbf{u})$. In addition, the van der Waals interaction potential $U(u_3)$ is approximated by [7,8,14]

$$U(\mathbf{u} \cdot \mathbf{e}_3) \approx -U_0 - F \mathbf{u} \cdot \mathbf{e}_3 - Y (\mathbf{u} \cdot \mathbf{e}_3)^2 / 2, \tag{4}$$

in which $U_0 = A/12\pi d^2$, $F = A/6\pi d^3$, and $Y = A/2\pi d^4$. Thus, minimization of the free energy (*i.e.* $\delta\Pi = 0$) demands $\nabla \cdot \boldsymbol{\sigma} = 0$, which, after substitution of (1) and (2), leads to

$$\begin{aligned} &\mu[\lambda^2 u_{1,11} + \lambda^{-1}(u_{1,22} + u_{1,33})] - p_{,1} = 0, \\ &\mu[\lambda^2 u_{2,11} + \lambda^{-1}(u_{2,22} + u_{2,33})] - p_{,2} = 0, \\ &\mu[\lambda^2 u_{3,11} + \lambda^{-1}(u_{3,22} + u_{3,33})] - p_{,3} = 0. \end{aligned} \tag{5}$$

The associated boundary conditions are derived as $\mathbf{u} = 0$ at $x_3 = -h$ and $\boldsymbol{\sigma} \cdot \mathbf{e}_3 = \gamma(u_{3,11} + u_{3,22})\mathbf{e}_3 + F\mathbf{e}_3 + Y u_3 \mathbf{e}_3$ on the film surface.

Obviously, the above boundary value problem has a homogeneous solution $u_i^{(0)} = 0$ and $p^{(0)} = -F + \mu\lambda^{-1}$. To examine the stability of the film, we search for another solution $u_i = u_i^{(0)} + u_i^{(1)}$ and $p = p^{(0)} + p^{(1)}$ in the vicinity of the homogeneous solution, in which $u_i^{(1)}$ and $p^{(1)}$ are small fluctuations of $u_i^{(0)}$ and $p^{(0)}$, respectively. Owing to the incompressibility, $u_i^{(1)}$ can be represented by $u_1^{(1)} = -\phi_{2,3}$, $u_2^{(1)} = \phi_{1,3}$ and $u_3^{(1)} = \phi_{2,1} - \phi_{1,2}$, with ϕ_1 and ϕ_2 being stream functions. We assume that ϕ_1, ϕ_2 and $p^{(1)}$

are of the periodic forms as

$$\begin{aligned}\phi_1 &= \Phi_1(x_3)e^{i(k_1x_1+k_2x_2)},\\ \phi_2 &= \Phi_2(x_3)e^{i(k_1x_1+k_2x_2)},\\ p^{(1)} &= P(x_3)e^{i(k_1x_1+k_2x_2)},\end{aligned} \tag{6}$$

in which $i=\sqrt{-1}$, and k_1 and k_2 are wave numbers per unit length in the x_1- and x_2-directions, respectively. In this situation, the equations in (5) become

$$\begin{aligned}&\mu(\Phi_{1,333}-l^2\Phi_{1,3}) - i\lambda k_2 P = 0,\\ &\mu(\Phi_{2,333}-l^2\Phi_{2,3}) + i\lambda k_1 P = 0,\\ &\mu(\Phi_{,33}-l^2\Phi) + i\lambda P_{,3} = 0,\end{aligned} \tag{7}$$

where $\Phi = k_2\Phi_1 + k_1\Phi_2$ and $l=\sqrt{\lambda^3 k_1^2 + k_2^2}$. The solution reads

$$\begin{aligned}\Phi_1 &= -c_1e^{-lx_3} + c_2e^{lx_3} + ik_2(c_5e^{-kx_3} - c_6e^{kx_3}) + c_7,\\ \Phi_2 &= -c_3e^{-lx_3} + c_4e^{lx_3} - ik_1(c_5e^{-kx_3} - c_6e^{kx_3}) + c_8,\\ P &= a(c_5e^{-kx_3} + c_6e^{kx_3}),\end{aligned} \tag{8}$$

with $a=\mu(\lambda^2-\lambda^{-1})kk_1^2$ and $k=\sqrt{k_1^2+k_2^2}$. Note that among the eight constants c_1, c_2,… and c_8 there exists the relation $k_2c_7 - k_1c_8 = 0$, and both c_7 and c_8 do not appear in the expressions of the displacement and stress. Therefore, imposing the boundary conditions at the film/substrate interface and on the film surface, we obtain six homogeneous linear algebraic equations about the six constants c_1, c_2,… and c_6. To ensure the existence of nontrivial solutions, the determinant of the coefficient matrix must vanish, yielding

$$Y = k^2\gamma + \frac{2\mu l\left[(k^2+l^2)f(k,l) - k^4 - 6k^2l^2 - l^4\right]}{\lambda k(k^2-l^2)g(k,l)}, \tag{9}$$

in which the functions $f(k,l)$ and $g(k,l)$ are given by

$$\begin{aligned}f(k,l) &= (k-l)^2\cosh[(k+l)h] + (k+l)^2\cosh[(k-l)h],\\ g(k,l) &= (k-l)\sinh[(k+l)h] - (k+l)\sinh[(k-l)h].\end{aligned} \tag{10}$$

Equation (7) provides a criterion for the stability of the pre-tensioned film: the film buckles if any positive root of k_1 and k_2 exists for a fixed value of Y. The parameter Y, increases drastically with decreasing the gap between the contactor and film surface, is effectively a controlling factor responsible for buckling occurrence. In general, Y attains the minimum Y_c at $k_1 = k_1^c$ and $k_2 = k_2^c$, where the critical wave numbers k_1^c and k_2^c are determined by $\partial Y/\partial k_1 = 0$ and $\partial Y/\partial k_2 = 0$. The magnitude of Y_c represents the threshold of buckling, while the wave vector $\mathbf{k} = k_1\mathbf{e}_1 + k_2\mathbf{e}_2$ characterizes the buckling mode. From eq. (9) it is seen that the effect of surface energy is incorporated only in the first term on the right side. The term is positive, plays the role of stabilizing the film surface [7–15].

We note that eq. (9) can be reduced to the result for an incompressible film without pre-tension in the limit of $\lambda\to 1$ [14], although $\lambda > 1$ has been assumed in the foregoing derivation. In that circumstance, due to $l = k$, only one parameter k appears in the expression of Y. If the effect of surface energy is neglected, we can show that the critical wave number is determined solely by the initial film thickness H, while the buckling threshold depends on both H and the shear modulus μ of the film. The explicit results read $k_c = 2.12/H$ and $Y_c = 6.22\mu/H$, which are exactly the same as those reported previously [7,8,14]. Since the wave vector $\mathbf{k}$ has no a preferential orientation, the corresponding buckling pattern lacks a long-range order.

However, the situation changes remarkably when the film is pre-tensioned, because the symmetry of the deformation is broken. For different values of the principal

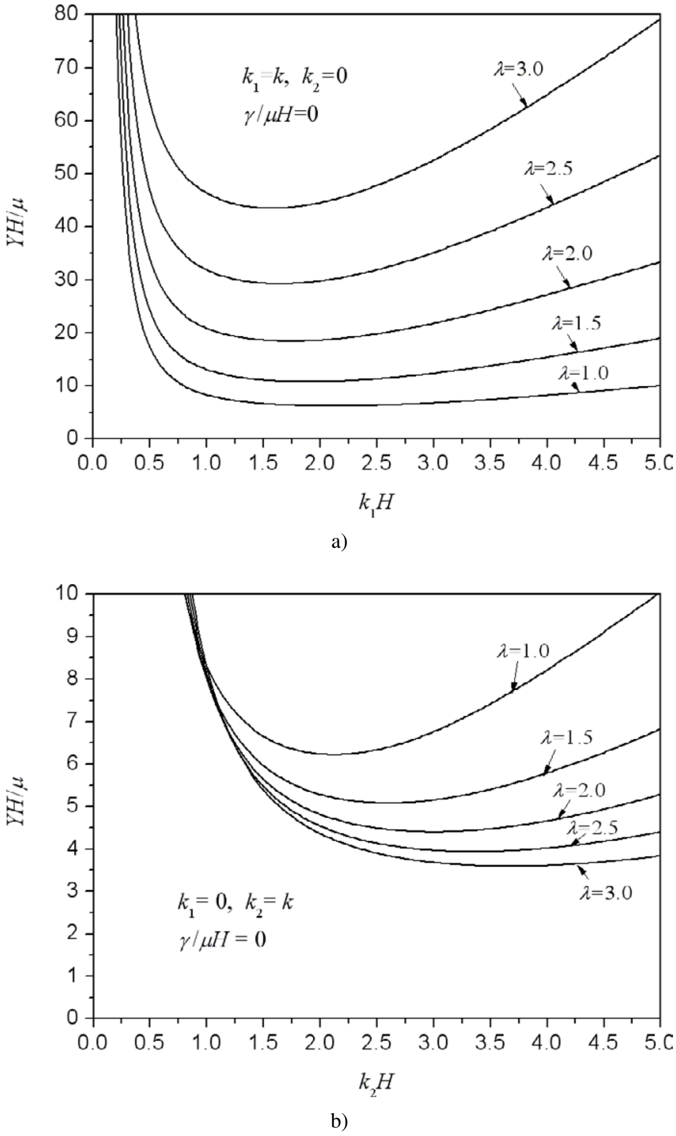

Fig. 2: Influence of the principal stretch λ on the dimensionless parameter YH/μ in two special buckling modes: a) $\mathbf{k} = k_1\mathbf{e}_1$ and b) $\mathbf{k} = k_2\mathbf{e}_2$. In both cases the normalized surface energy $\gamma/\mu H$ is assumed vanishing. For fixed initial thickness H and shear modulus μ of the film, the minimum of Y increases in case a) while decreases in case b) with λ increasing from 1.0 to 3.0.

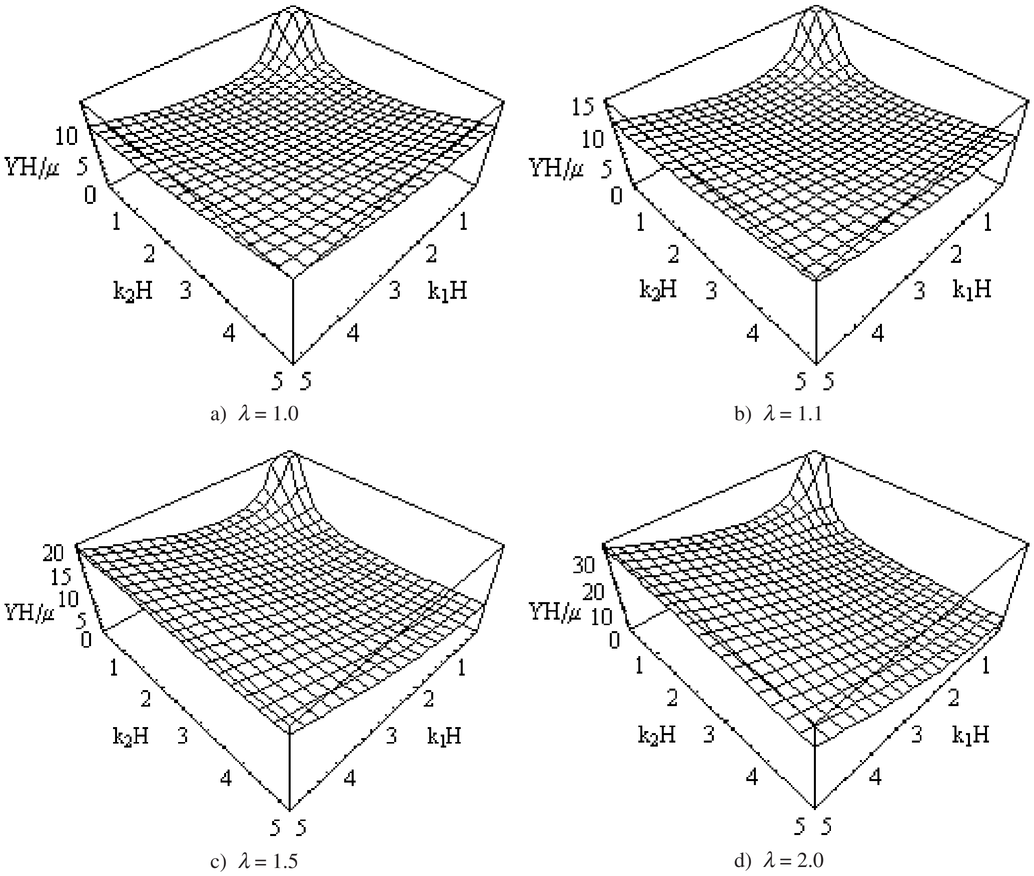

Fig. 3: Three-dimensional plots of the normalized Y-surface for different values of principal stretch λ: a) $\lambda=1.0$, b) $\lambda=1.1$, c) $\lambda=1.5$, and d) $\lambda=2.0$. The effect of surface energy is omitted. In case a) the surface possesses a rotational symmetry about the vertical axis, but in cases b), c), and d) the surfaces are wrapped, with the lowest points always located at the curve defined by $k_1=0$. The lager is λ, the lower is the position of the lowest point.

stretch λ, fig. 2 compares the results of the dimensionless parameter YH/μ calculated for two special modes of $\mathbf{k}=k_1\mathbf{e}_1$ and $\mathbf{k}=k_2\mathbf{e}_2$, where the effect of surface energy is omitted. We find that with increasing λ the magnitude of Y_c increases in the mode $\mathbf{k}=k_1\mathbf{e}_1$ while decreases in the mode $\mathbf{k}=k_2\mathbf{e}_2$. This implies that the pre-tension tends to inhibit the buckling in the former mode but facilitate the buckling in the latter mode. Unfortunately, from eq. (9) we are not able to obtain analytical solutions of the critical wave numbers. But an asymptotic expansion reveals that for $\lambda>1$ the relation in eq. (9) can be expressed by

$$Y = k^2\gamma + \frac{4\mu k[\cosh^2(kh)+k^2h^2]}{\lambda[\sinh(kh)-2kh]} + A(k)(l-k) + B(k)(l-k)^2 + \dots, \qquad (11)$$

where $A(k)$ and $B(k)$ are lengthy expressions in terms of k, H, λ, γ and μ. It can be shown that $A(k)$ and $B(k)$ are positive, thus for fixed k the parameter Y reaches the minimum Y_c at $k=l$, or equivalently at $k_1=0$. Indeed, this is clearly visible from the three-dimensional plots of Y as a function of k_1 and k_2 as given in fig. 3. Depicted in fig. 3a is the case of $\lambda=1.0$ (without pre-tension). In accordance with the foregoing analysis, the resulting Y-surface is a part of a revolution surface with the vale along the arc determined by $\sqrt{k_1^2+k_2^2}=k_c$. Figures 3b, c and d illustrate the cases for $\lambda=1.1$, 1.5 and 2.0, respectively. It is seen that the pre-tension wraps the Y-surfaces, and the lowest point on each surface always appears on the curve defined by $k_1=0$. The larger the principal stretch λ, the lower is the position of the lowest point. Therefore, we conclude that for $\lambda>1$ the critical buckling mode of the film must be $\mathbf{k}_c=k_2^c\mathbf{e}_2$, meaning that the film will buckle into stripes parallel to the tensile direction once the contactor is brought close enough to the film so that Y reaches the threshold Y_c. The magnitude of k_2^c can be obtained numerically. For different values of λ, the results of k_2^c and the corresponding Y_c are given in figs. 4 and 5, respectively. Evidently the critical wave number k_2^c is tunable by varying the principal stretch λ. In addition, the surface energy decreases k_2^c but increases Y_c, but it does not alter the buckling pattern substantially.

In summary, we have studied the effect of pre-tension on the buckling of a soft elastic film due to van der Waals interaction with a rigid contactor. The main conclusion is that a uni-axial pre-tension leads to striped buckling

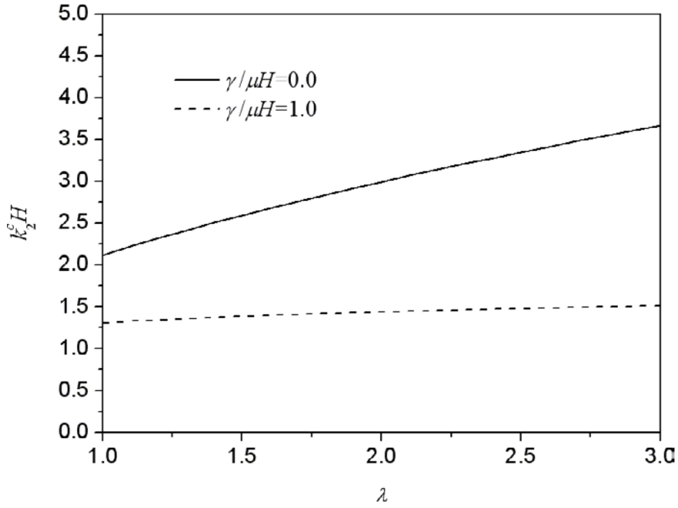

Fig. 4: Variations of the normalized critical wave number $k_2^c H$ with the principal stretch λ for different values of the normalized surface energy $\gamma/\mu H$. At the same value of λ, the normalized critical wave number for $\gamma/\mu H = 0.0$ (solid line) is larger than that for $\gamma/\mu H = 1.0$ (dashed line).

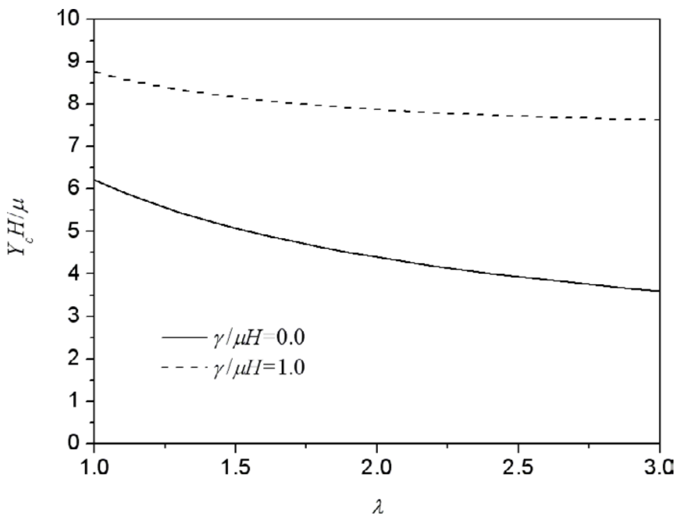

Fig. 5: Variations of the normalized buckling threshold $Y_c H/\mu$ with the principal stretch λ for different values of the normalized surface energy $\gamma/\mu H$. At the same value of λ, the normalized buckling threshold for $\gamma/\mu H = 0.0$ (solid line) is smaller than that for $\gamma/\mu H = 1.0$ (dashed line).

mode parallel to the tensile direction, and thus can be utilized to regulate the buckling pattern of the film. The result may find applications in the realms where precise surface patterns are desired, such as in the technologies of soft lithography and atomic force microscopy-assisted electrostatic lithography.

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