



# Surface wrinkling of a hyperelastic half-space coated by a liquid crystal elastomer film

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## ABSTRACT

We consider the stability of a hyperelastic substrate coated by a liquid crystal elastomer film and subjected to compressive forces. In this problem, the liquid crystal elastomer directors are free to evolve and this possible variation needs to be included in the stability analysis. We consider the case where the initial directors are aligned either in the horizontal or in the vertical direction and obtain an exact bifurcation condition for surface wrinkling. We show that director reorientation increases both the critical compressive strain and the critical wavenumber, hence stabilizing the material. In the small wavenumber limit we carry out an asymptotic analysis and obtain analytical solutions for the critical stretch and the critical wavenumber, which can be useful in applications.

## 1. Introduction

Liquid crystal elastomers (LCEs) combine the elasticity of rubber-like materials with the control features of liquid crystals (Warner and Terentjev, 2007). Depending on the arrangement of liquid crystal molecules, LCEs are classified as being in a nematic phase, smectic phase, or cholesteric phase. The nematic phase is the simplest since the director can be encoded by a single unit vector  $\mathbf{n}$ . Due to their fast and controllable reactions in response to various stimuli, such as mechanical loads (Conti et al., 2002; Ware et al., 2016), light (Sonnet and Virga, 2022; Liu et al., 2022), magnetic and electric fields (Urayama et al., 2005, 2006; Xu et al., 2020; Liu et al., 2020b), and heating (Agrawal et al., 2014; Wang et al., 2017b), LCEs have been suggested as promising materials in many applications such as artificial muscles (de Gennes et al., 1997; Tian et al., 2018), soft robotics (Zeng et al., 2018; DeSimone et al., 2015; Zhao et al., 2022), soft brakes (Wang et al., 2019b; Pang et al., 2019), and 4D printing (Wang et al., 2022a; Chen et al., 2023). An excellent starting point for the interested reader is the monograph by Warner and Terentjev (2007).

Many interesting features of LCEs were unraveled through experiments (see White and Broer (2015) and Xiao et al. (2023) for background). Mitchell et al. (1993) observed that the director of an LCE sample rotates in the loading direction. Kundler and Finkelmann (1995) discovered the “striping phenomenon” in a uniaxial stretching experiment of LCEs which Fried and Sellers (2006) showed to be fundamental and independent of the material softness. Under light or temperature stimuli, bending, curling and rolling of LCE specimens can be

induced (Potekhin and Wang, 2021; Zhai et al., 2021). Further, an indentation on flat LCE samples will create an elastic bulging (Mihai et al., 2023a).

In order to theoretically describe the deformation behavior of LCEs, a constitutive model is needed (Ball and Bedford, 2015). Accordingly, various strain–energy functions for LCEs have been suggested. Warner et al. (1988) extended the classical Gaussian theory of rubber elasticity to obtain the *neo-classical strain–energy function* that describes the deformation of LCEs under mechanical loads. Anderson et al. (1999) developed a continuum theory of LCEs where the strain–energy function is dependent on the deformation gradient, the director, and the gradient of director. Zhang et al. (2019) proposed a general theory applicable to the dynamics of LCEs with large deformations, director rotations and an order parameter that depends on temperature and light. Mihai et al. (2022) extended the classical Ogden model for rubber to study the auxetic responses of stretch deformation of LCEs. Recently, Wang et al. (2022c) developed a constitutive model to characterize viscoelastic behaviors of LCEs. Wu et al. (2023) presented a model to include the effect of loading-history dependence. Lee et al. (2023) proposed a macroscopic constitutive relation for polydomain LCEs. Since LCE samples are usually thin, there are many instances where they can be adequately modeled as plates. Indeed, the LCE samples made by Yan et al. (2012), Wang et al. (2017a, 2019a), Qing et al. (2019) are a few hundred microns in thickness. For this geometry, Krieger and Dias (2019) derived a von Kármán-type plate theory for LCEs and Mihai

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and Goriely (2020b) developed a new plate theory by decomposing the deformation gradient into elastic and natural deformations, which allows for the consideration of both optical and mechanical loads. Liu et al. (2020a) proposed a consistent plate model for LCEs subjected to mechanical loads suitable for finite deformations and without specific restriction on the applied load. Lately, Goriely et al. (2023) derived an LCE rod model suitable to describe illumination-induced deformations.

In general, the existence of directors and their deformations poses great challenges in solving the boundary value problems for such system. A way to bypass this problem is to carry out numerical simulations by modeling LCEs as a thermally sensitive material with anisotropic thermal expansions. Therefore, many commercial softwares can be directly used to study the mechanical behavior of LCEs (Mbanga et al., 2010; Zhao et al., 2023; Peng et al., 2023). More recently, the effect of director was also included in such codes (Chen et al., 2022). Further, Mahardika et al. (2024) performed a molecular dynamics simulation of deformations of LCEs under temperature effect.

As LCEs are typically thin, it is usual to support an LCE film by an elastic substrate, which creates a film–substrate bilayer. The surface wrinkling of bilayers has been of great interest in the last decades as it represents a fundamental instability in soft matter mechanics with a myriad of applications (Li et al., 2012; Goriely, 2017). An effective way to investigate such instabilities is to use the theory of incremental deformations, referred to as the small-on-large theory or incremental theory (Biot, 1965; Ogden, 1984). Within the framework of finite elasticity, Cai and Fu (1999) obtained a critical condition for the onset of surface wrinkling as well as the evolution of post-buckling of film/substrate structures. Since then, the effect of curvature (Jia et al., 2018), pre-stretch (Cao and Hutchinson, 2012), growth (Budday et al., 2015; Alawiye et al., 2019; Jin et al., 2019; Liu et al., 2021a), fibers (Stewart et al., 2016) were studied using theoretical, numerical, and experimental techniques. For LCE structures, Agrawal et al. (2012) prepared a bilayer system where a thin polystyrene film was attached to an LCE substrate and observed its wrinkling under temperature change using optical microscopy while Soni et al. (2016) studied theoretically surface wrinkling in an elastic film bonded to an LCE substrate. Fu et al. (2018) explored the surface instability of LCE film/substrate systems under light loading and applied Fourier spectral methods to investigate the post-buckling evolution. Krieger and Dias (2019) investigated surface wrinkling of a substrate coated by an LCE film using a plate model. Zhao et al. (2019) completed a finite element simulation for controllable surface patterns on LCE film–substrate structures by laser illumination. Xu and Zhao (2020) performed a finite element simulation on thermal wrinkling of LCE shell/core systems and studied the influence of director alignment on pattern selection. Starting from finite elasticity, Goriely and Mihai (2021) carried out a bifurcation analysis of a bilayer under biaxial compression where either the film or the substrate can be composed of LCEs. In particular, asymptotic solutions of the critical stretch triggering surface wrinkling as well as the critical wavenumber were deduced. In this model the director is assumed to be anchored. Barnes et al. (2023) conducted a systematic exploration on surface instabilities of LCE film–substrate structure generated in heating or cooling processes, and both experiments and numerical simulations were performed. In general, rotations of the director are possible which motivates the current study.

Here, we consider the coupling effect of director rotation of LCEs and the mechanical deformation induced by external loads. We develop a general incremental theory for LCEs within the framework of finite elasticity. Then, we conduct a standard bifurcation analysis to study the influence of various parameters on the onset and pattern obtained during wrinkling.

## 2. Problem formulation

We consider an LCE film bonded to a hyperelastic incompressible half-space. The thickness of the film in the initial configuration  $B_0$  is denoted by  $H$ . We assume that the bilayer undergoes a plane-strain deformation generated by a uniform compression, and as a result the thickness of the film becomes  $h$  in the current configuration  $B_1$ . As the compressive stress reaches a critical value, an instability takes place resulting in surface wrinkling (see Fig. 1).

We model the LCE film as an incompressible materials with a director field (Warner et al., 1988; Bladon et al., 1993; Anderson et al., 1999). Therefore, our problem is the generalization of the classical elastic incompressible bilayer problem under compression to the case where the film also includes directors. To distinguish between the LCE film and the substrate, we refer to any quantities related to the substrate by a hat “ $\hat{\phantom{x}}$ ”, and without a hat for the LCE film. For example, the strain-energy function of the LCE layer is written as  $W$  and the counterpart of the substrate is represented by  $\hat{W}$ .

For our problem, it is convenient to use a Cartesian coordinate system in both the reference and the current configurations. Accordingly, the position vectors of a material point in  $B_0$  and  $B_1$  are denoted by  $\mathbf{X} = (X_1, X_2)$  and  $\mathbf{x} = (x_1, x_2)$ , respectively. The same orthonormal basis  $\{\mathbf{E}_1, \mathbf{E}_2\}$  in all configurations will be used.

For the LCE film, we adopt the neo-classical constitutive model proposed by Warner et al. (1988) with an equivalent strain-energy function for isothermal deformation (Bladon et al., 1993; Fried and Korchagin, 2002):

$$W = \frac{\mu}{2} \left( \text{tr}(\mathbf{F}\mathbf{F}^T) - \frac{s-1}{s} \mathbf{F}^T \mathbf{n} \cdot \mathbf{F}^T \mathbf{n} + (s-1) \mathbf{F} \mathbf{n}_0 \cdot \mathbf{F} \mathbf{n}_0 - \frac{(s-1)^2}{s} (\mathbf{F}^T \mathbf{n} \cdot \mathbf{n}_0)^2 - 3 \right), \quad (2.1)$$

where  $\mu$  denotes the ground-state shear modulus,  $\mathbf{F}$  is the deformation gradient,  $\mathbf{n}_0$  and  $\mathbf{n}$  are the directors in the reference and the deformed configurations, respectively,  $s$  is a positive parameter measuring anisotropy, and the superscript “T” stands for transpose. When  $s = 1$ , (2.1) reduces to the energy density of the neo-Hookean model. The liquid crystal molecules are oblate if  $s < 1$  and prolate if  $s > 1$ . We further assume that the directors  $\mathbf{n}_0$  and  $\mathbf{n}$  are both unit vectors that are in the  $(X_1, X_2)$ -plane.

For the substrate, we use the incompressible neo-Hookean model:

$$\hat{W} = \frac{\hat{\mu}}{2} \left( \text{tr}(\hat{\mathbf{F}}\hat{\mathbf{F}}^T) - 3 \right), \quad (2.2)$$

where  $\hat{\mu}$  represents the ground-state shear modulus of the substrate.

The nominal stress tensors of the LCE film and of the substrate are given by

$$\mathbf{S} = \frac{\partial W(\mathbf{F}, \mathbf{n})}{\partial \mathbf{F}} - p \mathbf{F}^{-1}, \quad \hat{\mathbf{S}} = \frac{\partial \hat{W}(\hat{\mathbf{F}})}{\partial \hat{\mathbf{F}}} - \hat{p} \hat{\mathbf{F}}^{-1}, \quad (2.3)$$

where the pressures  $p$  and  $\hat{p}$  arise from the incompressibility constraints  $\det \mathbf{F} = 1$  and  $\det \hat{\mathbf{F}} = 1$ . An important difference with a typical elastic theory is the appearance of internal orientational body-force density for the LCE film given by

$$\mathbf{\Pi} = - \frac{\partial W(\mathbf{F}, \mathbf{n})}{\partial \mathbf{n}} - p_1 \mathbf{n}, \quad (2.4)$$

where  $p_1$  is the Lagrange multiplier enforcing the constraint  $\mathbf{n} \cdot \mathbf{n} = 1$ .

### 2.1. Homogeneous deformation

We first study the homogeneous deformation and characterize the current finite-deformation state  $B_1$ . All physical quantities associated with  $B_1$  will be added on an overbar “ $\bar{\phantom{x}}$ ”, except for the position vector  $\mathbf{x}$ . In view of the fact that both layers are incompressible, i.e.  $\det \bar{\mathbf{F}} = 1$ , the deformation gradient valid for both the LCE film and the half-space reads

$$\bar{\mathbf{F}} = \begin{pmatrix} \lambda & 0 \\ 0 & 1/\lambda \end{pmatrix}, \quad (2.5)$$

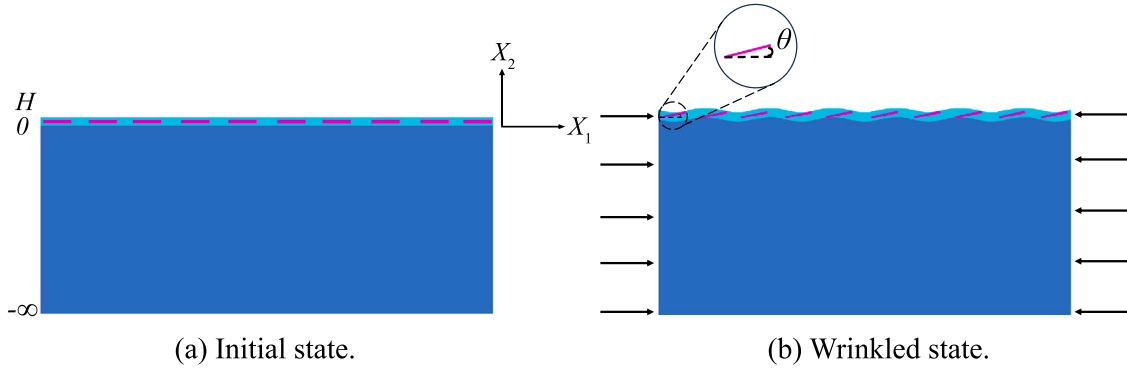


Fig. 1. Illustration of an LCE-substrate structure. (a) Initial state, where no force is applied to the structure; (b) Wrinkled state, where surface wrinkling is created by applied axial compression.

where  $\lambda$  is a constant principal stretch in the  $X_1$ -direction. The counterpart in the  $X_2$ -direction is then  $1/\lambda$ . We immediately find that the deformed thickness  $h = H/\lambda$ . Whenever a quantity in the substrate and in the film take the same value, we will discard the upper hat to denote both.

For the director, we have

$$\mathbf{n}_0 = \cos \theta_0 \mathbf{E}_1 + \sin \theta_0 \mathbf{E}_2, \quad \bar{\mathbf{n}} = \cos \bar{\theta} \mathbf{E}_1 + \sin \bar{\theta} \mathbf{E}_2, \quad (2.6)$$

where  $\theta_0$  and  $\bar{\theta}$  denote the angles between the director and the horizontal direction  $\mathbf{E}_1$  in the initial configuration  $B_0$  and the reference configuration  $B_1$ , respectively.

The nominal stresses  $\bar{\mathbf{S}}$  and  $\hat{\mathbf{S}}$  are also constant tensors. Hence for a static problem, the two equilibrium equations

$$\text{Div } \bar{\mathbf{S}} = \mathbf{0}, \quad \text{Div } \hat{\mathbf{S}} = \mathbf{0} \quad (2.7)$$

are trivially satisfied. Note that “Div” is the divergence operator with respect to the coordinates in  $B_0$ . We further study the orientational momentum balance which results in the equation  $\bar{\mathbf{T}} = \mathbf{0}$  for the material constitutive model (2.1) (Anderson et al., 1999). Further, we emphasize that the multiplier arising from the constraint of the director is negligible as shown in Anderson et al. (1999) and Chen and Fried (2006). To eliminate  $p_1$  in (2.4), we apply a projection operator  $(\mathbf{I} - \bar{\mathbf{n}} \otimes \bar{\mathbf{n}})$  on  $\bar{\mathbf{T}} = \mathbf{0}$  to find (Liu et al., 2020a)

$$(\mathbf{I} - \bar{\mathbf{n}} \otimes \bar{\mathbf{n}}) \frac{\partial W(\bar{\mathbf{F}}, \bar{\mathbf{n}})}{\partial \bar{\mathbf{n}}} = \mathbf{0}. \quad (2.8)$$

In the light of (2.1), (2.5), and (2.6), Eq. (2.8) gives rise to the following scalar equation

$$(s-1) \cos 2\bar{\theta} \sin 2\theta_0 - \frac{\sin 2\bar{\theta} \left( (\lambda^4 + 1)(s-1) \cos 2\theta_0 + (\lambda^4 - 1)(s+1) \right)}{2\lambda^2} = 0. \quad (2.9)$$

We then immediately obtain

$$\tan 2\bar{\theta} = \frac{2(s-1)\lambda^2 \sin 2\theta_0}{(1+s)(\lambda^4 - 1) + (s-1)(\lambda^4 + 1) \cos 2\theta_0}. \quad (2.10)$$

It can be seen from (2.10) that if  $\theta_0 = 0$  or  $\theta_0 = \pi/2$  the only possible solution is  $\bar{\theta} = 0$  or  $\bar{\theta} = \pi/2$  (as we are only concerned with  $0 \leq \bar{\theta} \leq \pi/2$ ). It implies that a director reorientation may be triggered whenever the initial director is either parallel or perpendicular to the  $X_1$ -axis. We emphasize that such phenomenon was also discovered in compressed bilayers (Mihai and Goriely, 2021; Goriely and Mihai, 2021), in inflated LCE tubes (Chen and Fried, 2006), and in the bending of LCEs (Liu et al., 2021b). We refer to Mihai and Goriely (2021) for a detailed discussion on the parameter selections needed to avoid director reorientation.

In the following analysis, we focus on the special case where the initial angle is  $\theta_0 = 0$  ( $\mathbf{n}_0 = \mathbf{E}_1$ ) or  $\theta_0 = \pi/2$  ( $\mathbf{n}_0 = \mathbf{E}_2$ ). Consequently,

there are four homogeneous solutions characterized by:  $\{\theta_0 = 0, \bar{\theta} = 0\}$ ,  $\{\theta_0 = 0, \bar{\theta} = \pi/2\}$ ,  $\{\theta_0 = \pi/2, \bar{\theta} = 0\}$ , and  $\{\theta_0 = \pi/2, \bar{\theta} = \pi/2\}$ .

For these solutions, Eq. (2.3)<sub>1</sub> yields

$$\begin{aligned} \bar{\mathbf{S}} &= \begin{pmatrix} \lambda\mu - \frac{\bar{p}}{\lambda} & 0 \\ 0 & \frac{\mu}{\lambda} - \lambda\bar{p} \end{pmatrix}, \quad \text{for } \{\theta_0 = 0, \bar{\theta} = 0\} \text{ or } \{\theta_0 = \pi/2, \bar{\theta} = \pi/2\}, \\ \bar{\mathbf{S}} &= \begin{pmatrix} s\lambda\mu - \frac{\bar{p}}{\lambda} & 0 \\ 0 & \frac{\mu}{s\lambda} - \lambda\bar{p} \end{pmatrix}, \quad \text{for } \{\theta_0 = 0, \bar{\theta} = \pi/2\}, \\ \bar{\mathbf{S}} &= \begin{pmatrix} \frac{\lambda\mu}{s} - \frac{\bar{p}}{\lambda} & 0 \\ 0 & \frac{s\mu}{\lambda} - \lambda\bar{p} \end{pmatrix}, \quad \text{for } \{\theta_0 = \pi/2, \bar{\theta} = 0\}. \end{aligned} \quad (2.11)$$

Accordingly, the nominal stress for the substrate is

$$\hat{\mathbf{S}} = \begin{pmatrix} \lambda\hat{\mu} - \hat{p}/\lambda & 0 \\ 0 & \hat{\mu}/\lambda - \lambda\hat{p} \end{pmatrix}. \quad (2.12)$$

Next, we consider the boundary and interface conditions. We assume that the upper surface is traction-free and the interface remains bonded which implies that continuity conditions are imposed at the interface. For the homogeneous deformation, we have

$$\bar{S}_{22}|_{X_2=H} = 0, \quad (\bar{S}_{22} - \hat{S}_{22})|_{X_2=0} = 0, \quad (2.13)$$

from which we obtain

$$\begin{aligned} \hat{p} &= \frac{\hat{\mu}}{\lambda^2}, \quad \bar{p} = \frac{\mu}{\lambda^2} \quad (\{\theta_0 = 0, \bar{\theta} = 0\} \text{ or } \{\theta_0 = \pi/2, \bar{\theta} = \pi/2\}), \\ \bar{p} &= \frac{\mu}{s\lambda^2}, \quad (\{\theta_0 = 0, \bar{\theta} = \pi/2\}), \quad \bar{p} = \frac{s\mu}{\lambda^2}, \quad (\{\theta_0 = \pi/2, \bar{\theta} = 0\}). \end{aligned} \quad (2.14)$$

As there are two solutions  $\bar{\theta} = 0$  and  $\bar{\theta} = \pi/2$ , it is necessary to identify which is the favorable one by computing the total energy of each solution. We select a representative block area from the original structure with  $[-L, L] \times [0, H] \cup [-L, L] \times [-50H, 0]$ . The equivalence to the prototypical problem can be attained by applying periodic conditions at the two lateral sides. For a displacement-control problem, the dimensionless two-dimensional energy is

$$\begin{aligned} \Psi &= \frac{\Psi^*}{LH\hat{\mu}} \\ &= \frac{1}{LH\hat{\mu}} \left( \int_{-L}^L \int_0^H W dX_1 dX_2 + \int_{-L}^L \int_{-50H}^0 \hat{W} dX_1 dX_2 \right). \end{aligned} \quad (2.15)$$

When the director  $\mathbf{n}_0$  is initially aligned in the  $\mathbf{E}_1$ -direction, with the use of (2.1) and (2.2), the corresponding energy for the two solutions  $\bar{\theta} = 0$  and  $\bar{\theta} = \pi/2$  read

$$\Psi_0^0 = \frac{(\beta + 50)(\lambda^2 - 1)^2}{\lambda^2}, \quad \Psi_{\pi/2}^0 = \frac{50(\lambda^2 - 1)^2}{\lambda^2} + \beta \left( \lambda^2 s + \frac{1}{\lambda^2 s} - 2 \right), \quad (2.16)$$

where the superscript and subscript correspond to the initial and deformed directors, respectively, and  $\beta = \mu/\hat{\mu}$  is the modulus ratio between two layers, an important parameter in the wrinkling problem.

It follows from (2.16) that  $\Psi_0^0$  is lower than  $\Psi_{\pi/2}^0$  as  $\lambda > s^{-1/4}$ , and the only positive real solution to  $\Psi_0^0 = \Psi_{\pi/2}^0$  is  $\lambda = s^{-1/4}$ . Therefore, we conclude that a director reorientation (director jumps from  $\mathbf{E}_1$  to  $\mathbf{E}_2$ ) is possible only if  $s > 1$ . Furthermore, if the director  $\mathbf{n}_0$  is initially aligned in the  $\mathbf{E}_2$ -direction, we find that

$$\Psi_0^{\pi/2} = \frac{50(\lambda^2 - 1)^2}{\lambda^2} + \beta \left( \frac{\lambda^2}{s} + \frac{s}{\lambda^2} - 2 \right), \quad \Psi_{\pi/2}^{\pi/2} = \frac{(\beta + 50)(\lambda^2 - 1)^2}{\lambda^2}. \quad (2.17)$$

In this scenario ( $\mathbf{n}_0 = \mathbf{E}_2$ ), we find that a director reorientation (with a jump from  $\mathbf{E}_2$  to  $\mathbf{E}_1$ ) can only be initiated if  $s < 1$  and the critical compressive stretch is  $\lambda = s^{1/4}$ .

## 2.2. Incremental theory

The incremental theory for hyperelastic material is now well established and we refer the reader to the excellent description found in (Ogden, 1984; Fu and Ogden, 1999). Therefore, we shall not repeat all the steps here but, instead, we will follow the standard procedure to obtain the required equations and boundary conditions. Here we need to extend the fundamental theory to the case of LCE film for which we have the added complication introduced by the director field. Therefore, we will only include this novel aspect for the film while the counterpart for the substrate will be given without derivation.

We expect that a wrinkling instability will be triggered as the applied deformation increases. To characterize the onset of instability, we superimpose an incremental displacement  $\mathbf{u} = [u_1, u_2]^T$  and an incremental director  $\mathbf{m} = [m_1, m_2]^T$  on  $B_1$ . The resulting configuration is called  $B_2$ . The position vector for a representative material particle in  $B_2$  is represented by  $\tilde{\mathbf{x}}$  and the associated director by  $\tilde{\mathbf{n}}$ . We have

$$\mathbf{u}(x_1, x_2) = \tilde{\mathbf{x}}(x_1, x_2) - \mathbf{x}, \quad \mathbf{m}(x_1, x_2) = \tilde{\mathbf{n}}(x_1, x_2) - \mathbf{n}. \quad (2.18)$$

It then follows that the deformation gradient  $\mathbf{F}$  arising from  $B_0 \rightarrow B_2$  is

$$\mathbf{F} = (\mathbf{I} + \boldsymbol{\eta})\bar{\mathbf{F}}, \quad (2.19)$$

where  $\mathbf{I}$  is the second-order identity tensor and  $\boldsymbol{\eta}$  the displacement gradient tensor with components  $\eta_{ij} = u_{i,j}$ . Here, the comma denotes derivative with respect to the implied coordinates. Based on (2.18)<sub>2</sub>, the internal orientational body-force density in  $B_2$  (2.4) can be replaced by

$$\boldsymbol{\Pi} = -\frac{\partial W(\mathbf{F}, \mathbf{n})}{\partial \mathbf{n}} - p_1(\mathbf{m} + \tilde{\mathbf{n}}). \quad (2.20)$$

To obtain the incremental equations applicable to the LCE film, we first define the incremental nominal stress

$$\hat{\mathbf{S}} = \bar{\mathbf{J}}^{-1} \bar{\mathbf{F}}(\mathbf{S} - \bar{\mathbf{S}}), \quad (2.21)$$

where  $\bar{\mathbf{J}} = \det \bar{\mathbf{F}}$ , which is one in this problem but is still kept so that the formula can be easily generalized. Similarly, we define the internal orientational body-force density increment by

$$\hat{\boldsymbol{\Pi}} = \bar{\mathbf{J}}^{-1}(\boldsymbol{\Pi} - \bar{\boldsymbol{\Pi}}) = \bar{\mathbf{J}}^{-1} \left( -\frac{\partial W}{\partial \mathbf{n}} + \frac{\partial W}{\partial \tilde{\mathbf{n}}} - p_1^* \tilde{\mathbf{n}} - p_1 \mathbf{m} \right). \quad (2.22)$$

Note that we have used  $p_1^* = p_1 - \bar{p}_1$  in the above equation.

To eliminate the Lagrange multipliers generated by the constraint of director, we use the constraint  $\mathbf{n} \cdot \mathbf{n} = 1$  which gives

$$\mathbf{m} \cdot (2\tilde{\mathbf{n}} + \mathbf{m}) = 0. \quad (2.23)$$

Next, applying the two tensors  $\mathbf{I} - \mathbf{a} \otimes \mathbf{a}/(\mathbf{a} \cdot \mathbf{a})$  and  $\mathbf{I} - \mathbf{m} \otimes \mathbf{m}/(\mathbf{m} \cdot \mathbf{m})$  with  $\mathbf{a} = \tilde{\mathbf{n}} + \mathbf{m}/2$  (Liu et al., 2020a) and using (2.23) we obtain the

incremental equations:

$$\begin{aligned} \operatorname{div} \hat{\mathbf{S}} &= \mathbf{0}, \\ \left( \mathbf{I} - \mathbf{a} \otimes \frac{\mathbf{a}}{\mathbf{a} \cdot \mathbf{a}} \right) \left( \mathbf{I} - \mathbf{m} \otimes \frac{\mathbf{m}}{\mathbf{m} \cdot \mathbf{m}} \right) \hat{\boldsymbol{\Pi}} &= \mathbf{0}, \quad \text{with} \quad \hat{\boldsymbol{\Pi}} \equiv -\frac{\partial W}{\partial \mathbf{n}} + \frac{\partial W}{\partial \tilde{\mathbf{n}}}, \end{aligned} \quad (2.24)$$

where “div” denotes the divergence operator with respect to the coordinates in  $B_1$ .

We denote by  $p^*$  the incremental pressure for the LCE film such that  $p = \bar{p} + p^*$  in  $B_2$  and taking the series expansions of  $\hat{\mathbf{S}}$  and  $\hat{\boldsymbol{\Pi}}$  in terms of  $(\tilde{\mathbf{F}}, \tilde{\mathbf{n}})$  to first order, we obtain (Ogden, 1984; Fu and Ogden, 1999):

$$\begin{aligned} \hat{S}_{ji} &= \mathcal{A}_{jilk} u_{k,l} + \bar{p} u_{j,i} - p^* \delta_{ji} + \mathcal{B}_{jik} m_k, \\ \hat{\Pi}_k &= -\mathcal{B}_{ljk} u_{j,l} - \mathcal{C}_{kj} m_j, \end{aligned} \quad (2.25)$$

where  $\delta_{ji}$  is the Kronecker delta, and  $\mathcal{A}_{jilk}$ ,  $\mathcal{B}_{jik}$ ,  $\mathcal{C}_{ji}$  are the first-order instantaneous elastic moduli defined by

$$\begin{aligned} \mathcal{A}_{jilk} &= \bar{\mathbf{J}}^{-1} \bar{\mathbf{F}}_{jA} \bar{\mathbf{F}}_{lB} \frac{\partial^2 W}{\partial F_{iA} \partial F_{kB}} \Big|_{\mathbf{F}=\bar{\mathbf{F}}, \mathbf{n}=\bar{\mathbf{n}}}, \\ \mathcal{B}_{jik} &= \bar{\mathbf{J}}^{-1} \bar{\mathbf{F}}_{jA} \frac{\partial^2 W}{\partial F_{iA} \partial n_k} \Big|_{\mathbf{F}=\bar{\mathbf{F}}, \mathbf{n}=\bar{\mathbf{n}}}, \quad \mathcal{C}_{ji} = \bar{\mathbf{J}}^{-1} \frac{\partial^2 W}{\partial n_j \partial n_i} \Big|_{\mathbf{F}=\bar{\mathbf{F}}, \mathbf{n}=\bar{\mathbf{n}}}. \end{aligned} \quad (2.26)$$

Accordingly, the incremental equations (2.24) transform into

$$\begin{aligned} \mathcal{A}_{jilk} u_{k,lj} - p_i^* + \mathcal{B}_{jik} m_{k,j} &= 0, \\ (\delta_{ik} - \bar{n}_i \otimes \bar{n}_k)(-\mathcal{B}_{ljk} u_{j,l} - \mathcal{C}_{kj} m_j) &= 0. \end{aligned} \quad (2.27)$$

The linearized incompressibility condition and the unit director constraint are prescribed by

$$u_{i,i} = 0, \quad m_i \cdot \bar{n}_i = 0. \quad (2.28)$$

The incremental boundary conditions at the upper surface and continuity conditions at the interface lead to

$$\mathbf{T}|_{x_2=h} = \mathbf{0}, \quad (\mathbf{T} - \hat{\mathbf{T}})|_{x_2=0} = \mathbf{0}, \quad (\mathbf{u} - \hat{\mathbf{u}})|_{x_2=0} = \mathbf{0}, \quad (2.29)$$

where  $\mathbf{T} = \hat{\mathbf{S}}^T \mathbf{E}_2$  is the traction vector and  $\hat{\mathbf{T}}$  the counterpart for the substrate with components given by

$$\begin{aligned} T_i &= \mathcal{A}_{2ilk} u_{k,l} + \bar{p} u_{2,i} - p^* \delta_{2i} + \mathcal{B}_{2ik} m_k, \\ \hat{T}_i &= \mathcal{A}_{2ilk} \hat{u}_{k,l} + \hat{p} \hat{u}_{2,i} - \hat{p}^* \delta_{2i}. \end{aligned}$$

Finally, we write the incremental equations for the substrate and the associated decaying condition at infinity as

$$\begin{aligned} \hat{u}_{i,i} &= 0, \quad \mathcal{A}_{jilk} \hat{u}_{k,lj} - \hat{p}_{,i}^* = 0, \\ \hat{u}_i &\rightarrow 0, \quad \text{as } x_2 \rightarrow -\infty. \end{aligned} \quad (2.30)$$

## 3. Bifurcation analysis

We are now in a position to carry out a bifurcation analysis based on the incremental governing system (2.27)–(2.30). We will take  $\theta_0 = 0$  as an example. In this case, if there is no director reorientation ( $\bar{\theta} = 0$ ), (2.28)<sub>2</sub> gives rise to  $m_1 = 0$ . As a result, we look for a solution of the form

$$\begin{aligned} u_1 &= e^{ikx_1} k U'(kx_2), \quad u_2 = -ike^{ikx_1} U(kx_2), \\ \hat{u}_1 &= e^{ikx_1} k \hat{U}'(kx_2), \quad \hat{u}_2 = -ike^{ikx_1} \hat{U}(kx_2), \\ m_2 &= e^{ikx_1} V(x_2), \end{aligned} \quad (3.1)$$

where ‘i’ is the imaginary unit, primes denote derivative with respect to the implied variable,  $k$  stands for the wavenumber,  $U$ ,  $\hat{U}$  and  $V$  are unknown functions. A direct computation would show that the incompressibility condition is automatically satisfied by (3.1).

Substituting (3.1) into (2.27), (2.30)<sub>2</sub> and eliminating  $p^*$ ,  $\hat{p}^*$  through cross differentiation result in

$$\begin{aligned}
k^4 s^2 \lambda^4 U - k^2 (s-1) s \lambda^4 V - k^4 s (\lambda^4 + 1) U'' + k^2 (1-s) V'' + k^4 U'''' &= 0, \\
(s-1)(k^2 s \lambda^4 U + V + k^2 U'') &= 0, \\
\lambda^4 \hat{U} - (\lambda^4 + 1) \hat{U}'' + \hat{U}'''' &= 0.
\end{aligned} \tag{3.2}$$

We eliminate  $V$  in (3.2)<sub>1</sub> by using (3.2)<sub>2</sub>. As a result, the general solutions of  $U$  and  $\hat{U}$  can be formulated by solving the associated eigenvalue problems of (3.2)<sub>1</sub> and (3.2)<sub>3</sub>:

$$\begin{aligned}
U(kx_2) &= C_1 e^{-\gamma_1 kx_2} + C_2 e^{\gamma_1 kx_2} + C_3 e^{-\gamma_2 kx_2} + C_4 e^{\gamma_2 kx_2}, \\
\hat{U}(kx_2) &= C_5 e^{kx_2} + C_6 e^{\lambda^2 kx_2} + C_7 e^{-kx_2} + C_8 e^{-\lambda^2 kx_2},
\end{aligned} \tag{3.3}$$

where  $C_i$  ( $i = 1, 2, \dots, 8$ ) are constants and the eigenvalues  $\gamma_1$  and  $\gamma_2$  are given by

$$\begin{aligned}
\gamma_1 &= \left( \frac{1 + (3-2s)\lambda^4 - \sqrt{1 + (6-8s)\lambda^4 + (9-8s)\lambda^8}}{2} \right)^{1/2}, \\
\gamma_2 &= \left( \frac{1 + (3-2s)\lambda^4 + \sqrt{1 + (6-8s)\lambda^4 + (9-8s)\lambda^8}}{2} \right)^{1/2}.
\end{aligned}$$

The decaying condition (2.30)<sub>3</sub> leads to  $C_7 = C_8 = 0$ .

Subsequently, we substitute (3.3) into the boundary conditions and continuity conditions (2.29) to obtain an algebraic system of the general form

$$\mathbf{M}_1 \cdot \mathbf{C} = 0 \tag{3.4}$$

where  $\mathbf{C} = [C_1, C_2, C_3, C_4, C_5, C_6]^T$  and the coefficient matrix  $\mathbf{M}_1$  is given in (A.1). In order to find a non-trivial solution of the incremental displacements, the determinant of this matrix must vanish, resulting in the following bifurcation condition:

$$\det \mathbf{M}_1 \equiv f_1(\lambda, kh, \beta, s) = 0. \tag{3.5}$$

Since the bifurcation condition depends on  $k$  and  $h$  only via  $kh$ , we henceforth take  $kh$  as a single parameter.

From the previous section, we know that for some values of  $s > 1$ , the director will rotate from  $\mathbf{E}_1$  to  $\mathbf{E}_2$  at  $\lambda = s^{-1/4}$ . If this value is more than the critical stretch triggering surface wrinkling, we may expect a director rotation first and then a secondary instability. For this special case, (2.28)<sub>2</sub> yields  $m_2 = 0$  and therefore we shall seek a new increment  $m_1 = e^{ikx_1} V(x_2)$ . Accordingly, the general solution of the LCE film is replaced by

$$U(kx_2) = C_1 e^{-\gamma_3 kx_2} + C_2 e^{\gamma_3 kx_2} + C_3 e^{-\gamma_4 kx_2} + C_4 e^{\gamma_4 kx_2}, \tag{3.6}$$

where  $\gamma_3$  and  $\gamma_4$  are eigenvalues defined by

$$\begin{aligned}
\gamma_3 &= \left( \frac{s^2 \lambda^4 + 3 - 2s - (\lambda^8 s^4 - 8\lambda^4 s^3 + 6\lambda^4 s^2 - 8s + 9)^{1/2}}{2(s^2 \lambda^4 + s - 1)} \right)^{1/2} \lambda^2 s^{1/2}, \\
\gamma_4 &= \left( \frac{s^2 \lambda^4 + 3 - 2s + (\lambda^8 s^4 - 8\lambda^4 s^3 + 6\lambda^4 s^2 - 8s + 9)^{1/2}}{2(s^2 \lambda^4 + s - 1)} \right)^{1/2} \lambda^2 s^{1/2}.
\end{aligned}$$

Analogously, the bifurcation condition where surface instability occurs can be obtained from (2.29):

$$\det \mathbf{M}_2 \equiv f_2(\lambda, kh, \beta, s) = 0, \tag{3.7}$$

where  $\mathbf{M}_2$  is shown in (A.2).

For the case  $\theta_0 = \pi/2$ , there are also two possible situations, i.e.  $\bar{\theta} = \pi/2$  and  $\bar{\theta} = 0$ . The associated bifurcation conditions can be deduced by the same procedures and hence the details will be omitted. We directly write them as

$$\det \mathbf{M}_3 \equiv f_3(\lambda, kh, \beta, s) = 0, \quad \bar{\theta} = \frac{\pi}{2}, \tag{3.8}$$

$$\det \mathbf{M}_4 \equiv f_4(\lambda, kh, \beta, s) = 0, \quad \bar{\theta} = 0, \tag{3.9}$$

where  $\mathbf{M}_3$  and  $\mathbf{M}_4$  can be found in (A.3) and (A.4), respectively.

Using the bifurcation conditions (3.5) and (3.7)–(3.9), we can graph the bifurcation curves with varying  $kh$  as shown in Fig. 2. We see

that a director reorientation takes place before surface wrinkling when  $\theta_0 = 0, s = 2$  and  $\theta_0 = \pi/2, s = 0.8$  in Fig. 2(a) and when  $\theta_0 = \pi/2, s = 0.8$  in Fig. 2(b). In the first case ( $\beta = 1$ ), the LCE film and the substrate have the same modulus. When  $s = 1$ , the problem further reduces to a homogeneous half-space for which surface wrinkling occurs at the classical Biot value  $\lambda \approx 0.5437$  (Biot, 1963). Therefore, we refer to the critical stretch for surface instability when  $\beta = 1$  as  $\lambda_{\text{Biot}}$ . It can be seen from Fig. 2(a) that all bifurcation curves intersect when  $kh \rightarrow 0$  and the corresponding stretch is exactly at the Biot value 0.5437 (marked by a black point). In addition, if  $s < 1$  and  $\theta_0 = 0$  the principal stretch  $\lambda$  will converge to a limiting value as  $kh \rightarrow \infty$ , which is higher than 0.5437. Besides, the limit values as  $kh \rightarrow \infty$  are lower than 0.5437 for the rest of bifurcation curves, and hence the critical stretch is  $\lambda_{\text{Biot}} = 0.5437$ . Correspondingly, the associated critical wavenumber is zero. A zero wavenumber will typically cause a localized instability, such as ridge formation in a film bonded to a pre-compressed substrate (Fu et al., 2023) and localized necking/bulging in circular cylinders (Fu et al., 2021; Liu and Dorfmann, 2023). We further show  $\lambda_{\text{Biot}}$  as a function of  $s$  in Fig. 3. The blue curve is monotonically decreasing with  $s$  until the Biot value is reached at  $s = 1$  while the red one is a horizontal line. Moreover, the blue curve is always higher, indicating that the LCE-substrate bilayer is more stable when the director is initially along the  $\mathbf{E}_2$ -direction. Note that the director will reorientate if  $\theta_0 = \pi/2$  and  $s < 1$ . This further demonstrates that director reorientation stabilizes the material.

When the LCE film is stiffer than the substrate, as shown in Fig. 2(b) the  $\lambda - kh$  curve has a local maximum. We refer to this point as  $(\lambda_{\text{cr}}, (kh)_{\text{cr}})$  with critical stretch  $\lambda_{\text{cr}}$  and critical wavenumber  $(kh)_{\text{cr}}$ . Next, we shall focus on the effect of parameters on this critical pattern.

### 3.1. Critical wrinkled state if the initial director is aligned in the horizontal direction

We first specify  $\theta_0 = 0$  and use Eqs. (3.5) and (3.7) to study the critical pattern. The pair  $\{\lambda_{\text{cr}}, (kh)_{\text{cr}}\}$  can be determined from

$$f_i(\lambda, kh, \beta, s) = 0, \quad \frac{\partial f_i(\lambda, kh, \beta, s)}{\partial (kh)} = 0, \quad i = 1, 2. \tag{3.10}$$

We can solve these equations numerically for the critical stretch  $\lambda_{\text{cr}}$  and the critical wavenumber  $(kh)_{\text{cr}}$ . From (2.16) we know that a director jump from  $\bar{\theta} = 0$  to  $\bar{\theta} = \pi/2$  may take place at  $\lambda = s^{-1/4}$  when  $s > 1$ . Therefore, we study separately the cases  $s < 1$  and  $s > 1$ .

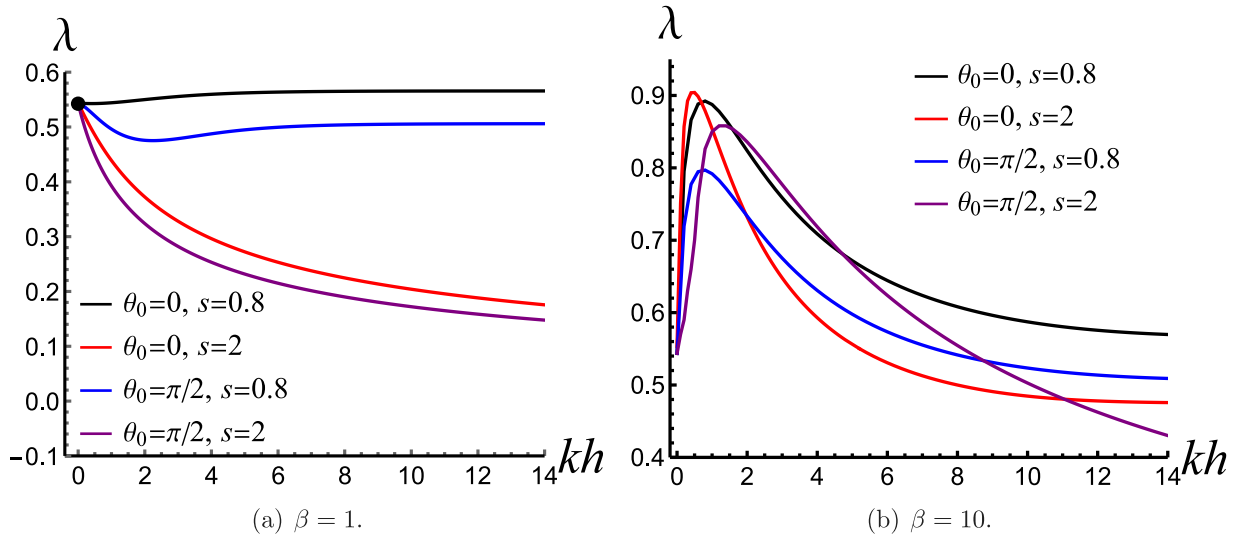
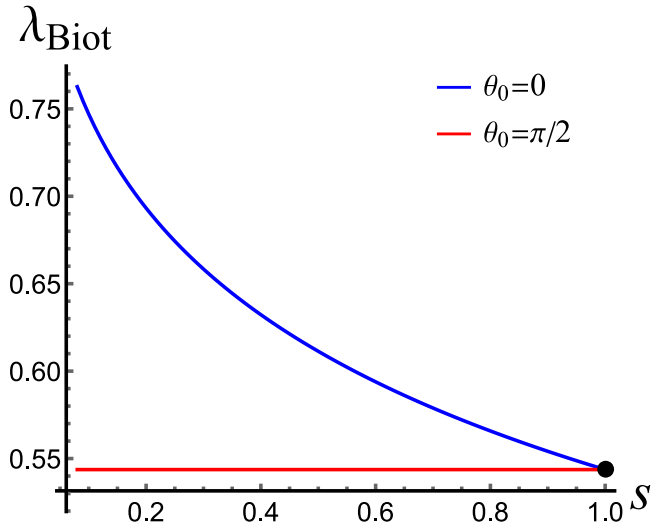
#### 3.1.1. The case of $s < 1$

First, we assume  $s < 1$  when the director remains in its initial direction before surface wrinkling. In that case, the critical stretch and the critical wavenumber are shown in Fig. 4 based on Eqs. (3.10) (with  $i = 1$ ). As can be appreciated from Fig. 4(a), when the LCE film is stiffer than the substrate, the critical stretch  $\lambda_{\text{cr}}$  increases with  $\beta$  regardless of the value of  $s$ , implying that the bilayer becomes more unstable as the modulus ratio  $\beta$  increases. This is a well-known property of film-substrate structures (Cai and Fu, 1999; Andres et al., 2018; Wang et al., 2023) which remains valid when a substrate is coated by an LCE film (Krieger and Dias, 2019; Goriely and Mihai, 2021). Meanwhile, the value of  $s$  has a weak influence on  $\lambda_{\text{cr}}$  provided that  $\beta$  is large enough, which has also been observed by Goriely and Mihai (2021). As  $\beta$  decreases, the curve will intersect with the horizontal line  $\lambda = \lambda_{\text{Biot}}$  and as a result the critical stretch will be replaced by  $\lambda_{\text{Biot}}$ .

We see from Fig. 4(b) that the critical wavenumber is a monotonically decreasing function of the modulus ratio  $\beta$ , regardless of the value of the anisotropy parameter  $s$ . This feature is also consistent with the results in Goriely and Mihai (2021). In particular, as  $\beta$  goes to one, the critical wavenumber  $(kh)_{\text{cr}}$  tends to infinity.

#### 3.1.2. The case of $s > 1$

Next, we consider  $s > 1$ . In this case, from (2.16), we know that a director reorientation can take place at  $\lambda = s^{-1/4}$ . Hence, director

Fig. 2. Bifurcation curves for  $\lambda$  as a function of  $kh$ .Fig. 3. The  $\lambda_{\text{Biot}}$  curves for  $\mathbf{n}_0 = \mathbf{E}_1$  (the blue curve) and  $\mathbf{n}_0 = \mathbf{E}_2$  (the red line) when  $s \leq 1$ .

reorientation will occur first before surface wrinkling if the critical stretch obtained from (3.10) (with  $i = 1$ ) is lower than  $s^{-1/4}$ . Since  $\lambda_{\text{cr}}$  is dependent on  $\beta$ , we need to identify the transition where a director reorientation is induced first instead of surface wrinkling. This can be achieved by substituting  $\lambda = s^{-1/4}$  into (3.10) (with  $i = 1$ ).

Fig. 5 shows that the critical modulus ratio  $\beta_{\text{cr}}$  depends monotonically on the anisotropy parameter  $s$ . We recall that there is no director reorientation provided  $\beta > \beta_{\text{cr}}$ . In practice, it provides a guidance to avoid or to trigger a director reorientation before surface wrinkling in the sample preparation process. For  $\beta < \beta_{\text{cr}}$ , the director will first rotate  $\pi/2$  at  $\lambda = s^{-1/4}$ , changing its alignment from  $\mathbf{E}_1$  to  $\mathbf{E}_2$ . In this case, we can use (3.10) (with  $i = 2$ ) to find  $\lambda_{\text{cr}}$  and  $(kh)_{\text{cr}}$ . Subsequently, we plot in Fig. 6 the curves of the critical stretch  $\lambda_{\text{cr}}$  and the critical wavenumber  $(kh)_{\text{cr}}$  against the modulus ratio  $\beta$  when  $s > 1$ . Due to the presence of director reorientation, all curves exhibit a discontinuity at  $\beta_{\text{cr}}$  and as a consequence a discontinuity in both  $\lambda_{\text{cr}}$  and  $(kh)_{\text{cr}}$ .

We see from Fig. 6(a) that director reorientation significantly delays wrinkling initiation. Moreover, as  $s$  increases, the magnitude of this delay effect also increases. When  $\beta > \beta_{\text{cr}}$ , the critical stretch is almost identical for different values of  $s$ . Notwithstanding, a careful observation reveals that  $\lambda_{\text{cr}}$  is an increasing function of  $s$ . If  $\beta < \beta_{\text{cr}}$ , the critical stretch decreases towards  $\lambda_{\text{Biot}}$ . Specifically, the blue curve, corresponding to  $s = 2$ , will reach  $\lambda_{\text{Biot}}$  after  $\beta$  crosses  $\beta_{\text{cr}}$ . Fig. 6(b) shows that the director reorientation prominently amplifies the critical wavenumber  $(kh)_{\text{cr}}$ . Also, the critical wavenumber  $(kh)_{\text{cr}}$  decreases in  $s$  until the termination of each curve. Referring to Fig. 2(a), it is expected that  $(kh)_{\text{cr}} = 0$  when  $\beta$  is approaching unity. This explains why these curves terminate as  $\beta$  is getting small. Especially, for the blue curve in Fig. 6(b),  $(kh)_{\text{cr}}$  will jump from a finite value to 0 immediately after  $\beta$  passes  $\beta_{\text{cr}}$ .

### 3.2. Critical wrinkled state if the initial director is aligned in the vertical direction

Next, we study the bifurcations for  $\theta_0 = \pi/2$ . From (2.17), we see that the director may rotate from  $\mathbf{E}_2$  to  $\mathbf{E}_1$  at  $\lambda = s^{1/4}$  if  $s < 1$ . Similarly, we will distinguish the cases  $s < 1$  and  $s > 1$ . As before, the pair  $\{\lambda_{\text{cr}}, (kh)_{\text{cr}}\}$  can be identified by

$$f_i(\lambda, kh, \beta, s) = 0, \quad \frac{\partial f_i(\lambda, kh, \beta, s)}{\partial(kh)} = 0, \quad i = 3, 4. \quad (3.11)$$

#### 3.2.1. The case of $s < 1$

In the case  $s < 1$ , we refer to the previous subsection and first determine the critical transition value  $\beta_{\text{cr}}$  where surface wrinkling will take place following a director reorientation. To this end, we directly substitute  $\lambda = s^{1/4}$  into Eqs. (3.11) (with  $i = 3$ ). The result is shown in Fig. 7, indicating that  $\beta_{\text{cr}}$  is a decreasing function of  $s$ . In particular, as  $s \approx 1/2$ , we find that  $\beta_{\text{cr}}$  is approaching zero, indicating that a further lower  $s$  fails to generate any director reorientation before surface wrinkling.

We show the curves of  $\lambda_{\text{cr}}$  and  $(kh)_{\text{cr}}$  in Fig. 8 for varying  $\beta$ . Fig. 8(a) illustrates that  $\lambda_{\text{cr}}$  slowly decreases with decreasing  $\beta$  until it passes the transition value  $\beta_{\text{cr}}$  where an obvious drop in  $\lambda_{\text{cr}}$  is observed. This again implies that director reorientation indeed retards surface wrinkling. After that point, the  $\lambda_{\text{cr}}$ -curve descends fast to  $\lambda_{\text{Biot}}$  as  $\beta$  further reduces. Fig. 8(b) shows that the critical wavenumber  $(kh)_{\text{cr}}$

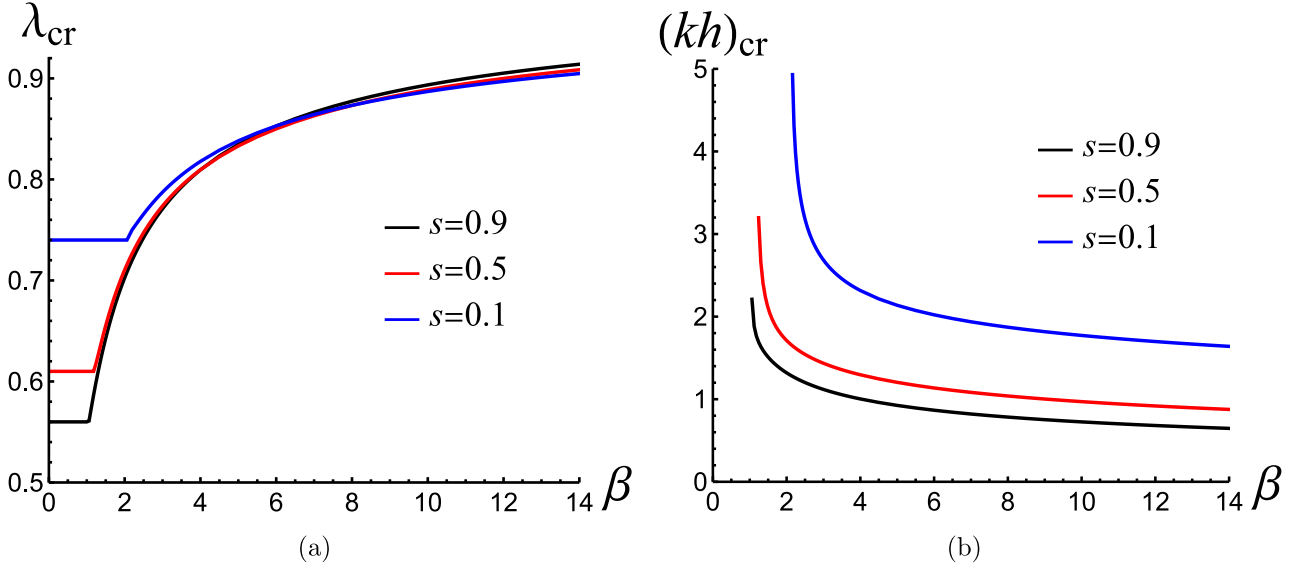


Fig. 4. The critical stretch  $\lambda_{cr}$  and the critical wavenumber  $(kh)_{cr}$  as functions of the modulus ratio  $\beta$  when  $\theta_0 = 0$  and  $s < 1$ .

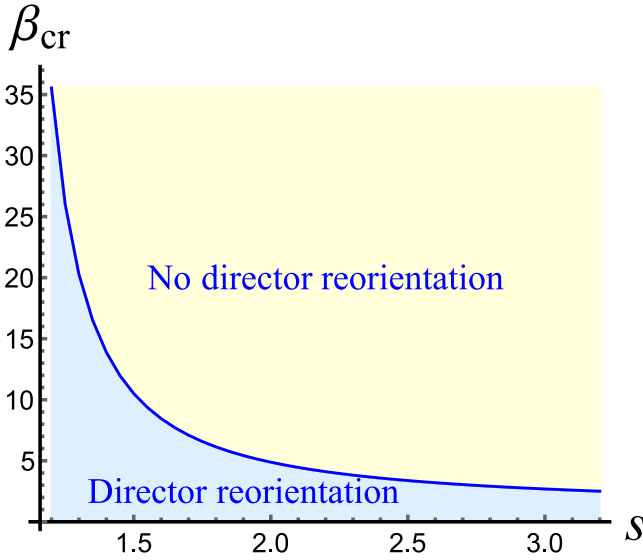


Fig. 5. Dependence of the critical modulus ratio  $\beta_{cr}$  on the anisotropy parameter  $s$  if  $\theta_0 = 0$  and  $s > 1$ .

depends marginally on the modulus ratio if  $\beta > \beta_{cr}$ . Once more, the  $(kh)_{cr}$  curve stops at a special value of  $\beta < \beta_{cr}$  and therefore we have  $(kh)_{cr} = 0$  since then. When director reorientation happens, all curves display a remarkable upper jump, leading to a great increase in the value of  $(kh)_{cr}$ . Combining the facts shown in Figs. 6 and 8, we conclude that a director reorientation actually significantly delays surface wrinkling while producing more wrinkles.

### 3.2.2. The case of $s > 1$

Next, we consider the case  $s > 1$  which excludes the possibility of director reorientation. In this case, we can directly obtain the exact solutions for  $\lambda_{cr}$  and  $(kh)_{cr}$  based on (3.11) (with  $i = 4$ ) and the results are displayed in Fig. 9. We find that  $\lambda_{cr}$  monotonically decreases to the Biot value 0.5437 (the horizontal line) as  $\beta$  decreases. However, the critical wavenumber  $(kh)_{cr}$  increases monotonically with reducing  $\beta$ .

### 3.3. The effect of the anisotropy parameter $s$

From Figs. 4, 6, 8 and 9, we find that the LCE film makes a great difference in affecting the critical state, especially when there is a director reorientation. As the anisotropy parameter  $s$  is critical for distinguishing LCEs and isotropic materials, we investigate the influence of  $s$  on  $\lambda_{cr}$  and  $(kh)_{cr}$  and present the results in Fig. 10. The discontinuities in all curves are caused by director reorientation. From the blue curve corresponding to  $\theta_0 = 0$ , we conclude that an LCE film always creates a more stable bilayer compared to its isotropic counterpart if  $s < 1$ . When  $1 < s < 1.3768$  (here the value 1.3768 can be found based on Fig. 5 by taking  $\beta = 15$ ), a sudden drop in  $\lambda_{cr}$  is observed and  $\lambda_{cr}$  reduces dramatically for increasing  $s$ . When  $s > 1.3768$ ,  $\lambda_{cr}$  is always higher than the critical stretch with  $s = 1$  and a greater  $s$  will make the bilayer unstable earlier. Meanwhile, the critical wavenumber  $(kh)_{cr}$  experiences a non-monotonic behavior due to director reorientation as  $s$  increases. When the initial director is aligned in the  $E_2$ -direction ( $\theta_0 = \pi/2$ ), the red curves imply that for  $0 < s < 0.7433$  (the value 0.7433 is identified based on Fig. 7 by taking  $\beta = 15$ ) we have an unstable bilayer compared to its isotropic counterpart ( $s = 1$ ). For  $0.7433 < s < 1$ , a director reorientation takes place and it retards the onset of surface wrinkling. When  $s > 1$ , the  $\lambda_{cr}$ -curve is monotonically decreasing in  $s$ , indicating that the bilayer is more stable.

In summary: A director reorientation will drastically delays surface wrinkling only if the anisotropy parameter  $s$  falls into a specific region, for example  $(0.7433, 1) \cup (1, 1.3768)$  in Fig. 10. Furthermore, if director reorientation is not granted, when  $s < 0.7433$ , the bilayer with  $\theta_0 = 0$  is always more stable than its counterpart with  $\theta_0 = \pi/2$ . However, if  $s > 1.3768$  the bilayer with  $\theta_0 = \pi/2$  is more stable. Recalling that prolate liquid crystal molecules imply  $s > 1$  while oblate molecules give  $s < 1$ , we conclude that when the major-axis of molecules coincides with the alignment of the director, a stable bilayer can be designed, and the associated wrinkled pattern is more wrinkled.

## 4. Asymptotic analysis

The exact bifurcation conditions (3.5) and (3.8) derived from the previous section are valid for all values of the geometric and material parameters. In practical applications, we are often in a regime with a

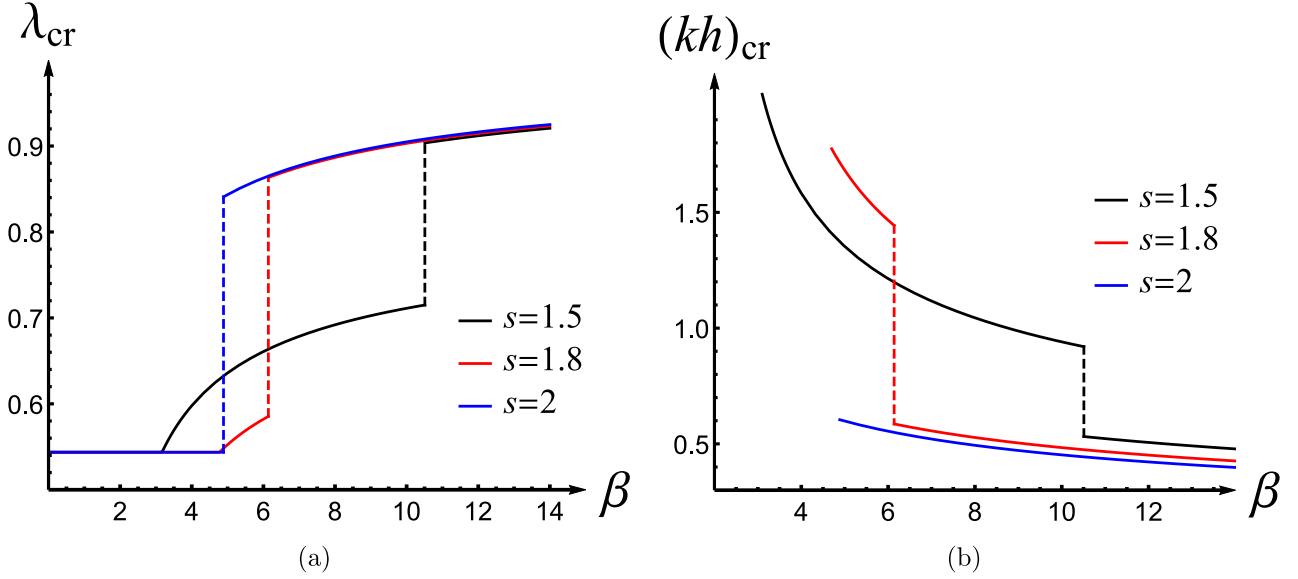


Fig. 6. The critical stretch  $\lambda_{cr}$  and the critical wavenumber  $(kh)_{cr}$  as functions of the modulus ratio  $\beta$  when  $\theta_0 = 0$  and  $s > 1$ .

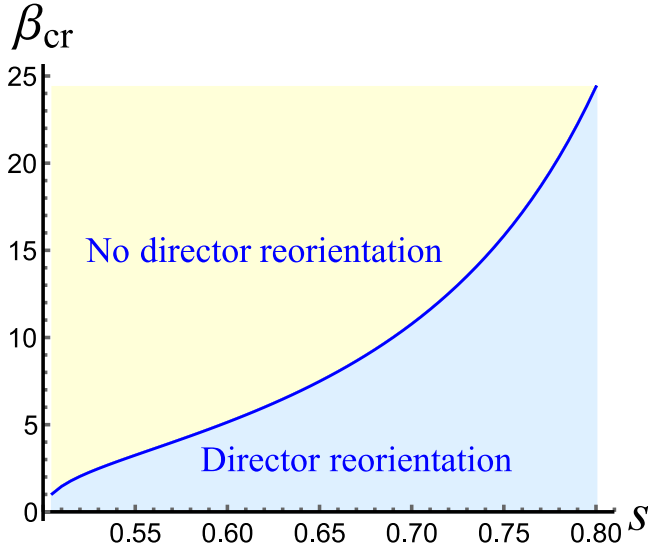


Fig. 7. Dependence of the critical modulus ratio  $\beta_{cr}$  on the anisotropy parameter  $s$  if  $\theta_0 = \pi/2$  and  $s < 1$ .

thick soft substrate coated by a stiff thin film (Cai and Fu, 1999; Li et al., 2012; Goriely, 2017; Holland et al., 2018). Hence, it is of importance to study the case  $\beta \gg 1$  in detail. In this regime an asymptotic analysis can also be performed. To this end, we further assume that  $kh \rightarrow 0$  and  $1/\beta \sim \mathcal{O}((kh)^3)$  (Cai and Fu, 1999; Liu and Dai, 2014; Alawiye et al., 2019; Goriely and Mihai, 2021).

From the analysis in Section 3 it is expected that, in the large  $\beta$  limit, surface wrinkling will take place when  $\lambda$  is close to 1. Further, it has also been shown in Sections 3.1 and 3.2 that a director reorientation will retard the onset of surface wrinkling and create more wrinkles. We emphasize that both effects violate the assumption of small  $kh$ . Thus, we restrict our analysis to the case where no director reorientation is allowed. Replacing  $\beta$  by  $\omega/(kh)^3$  with  $\omega$  a constant of  $\mathcal{O}(1)$ , we look for a solution to (3.5) and (3.8) depending only on the even orders of  $kh$

as follows (Goriely et al., 2008; De Pascalis et al., 2011; Liu, 2023)

$$\lambda = 1 + \zeta_1(kh)^2 + \zeta_2(kh)^4 + \mathcal{O}((kh)^6), \quad (4.1)$$

where  $\zeta_i$  ( $i = 1, 2$ ) are constants related to  $s$  and  $\omega$  to be determined. In the following, we will again study separately the two cases  $\theta_0 = 0$  and  $\theta_0 = \pi/2$ .

#### 4.1. The initial director is aligned in the horizontal direction

We first look for solutions when  $\theta_0 = 0$ . To avoid director reorientation, we impose  $s < 1$ . In addition, it can be concluded from Fig. 10(b) that  $kh$  is finite for small values of  $s$ . Therefore, we only consider  $s < 1$  and  $s \sim \mathcal{O}(1)$ . Using (4.1) and replacing  $\beta$  by  $\omega/(kh)^3$ , the bifurcation condition (3.5) depends on the small parameter  $kh$ . We expand (3.5) at each order in  $kh$  to find:

$$\begin{aligned} \zeta_1 &= -\frac{6 + s^2\omega}{12s\omega}, \\ \zeta_2 &= \frac{31s^5\omega^2 + 4s^4\omega(2\omega - 45) + 180s^3\omega - 900s + 1440}{1440s^3\omega^2}. \end{aligned} \quad (4.2)$$

Substituting  $\omega = \beta(kh)^3$  back into (4.2) gives rise to an expression for  $\lambda$  that solves the bifurcation condition (3.5):

$$\begin{aligned} \lambda &= 1 - \frac{1}{2(kh)s\beta} - \frac{(kh)^2s}{12} + \frac{(kh)^4s(31s + 8)}{1440} - \frac{(5s - 8)}{8(kh)^2s^3\beta^2} \\ &\quad - \frac{1}{8\beta}(kh)(s - 1) + \mathcal{O}((kh)^6). \end{aligned} \quad (4.3)$$

With the aid of (3.10)<sub>2</sub> with  $i = 1$  we obtain the asymptotic solutions for  $\lambda_{cr}$  and  $(kh)_{cr}$ :

$$\lambda_{cr} = 1 - \frac{3^{2/3}}{4s^{1/3}\beta^{2/3}} - \frac{3^{1/3}(4s + 7)(5s - 8)}{160s^{5/3}\beta^{4/3}} - \frac{3^{2/3}}{8s^{1/3}\beta^{5/3}} + \mathcal{O}(\beta^{-2}), \quad (4.4)$$

$$(kh)_{cr} = \frac{3^{1/3}}{s^{2/3}\beta^{1/3}} - \frac{(24 - 32s + 5s^2)}{20s^2\beta} + \mathcal{O}(\beta^{-5/3}). \quad (4.5)$$

It can be seen from (4.4) and (4.5) that, in addition to  $\beta$ , the anisotropy parameter  $s$  also appears at the leading-order, which explains both the delay to the bifurcation and the increase in wrinkles when  $s < 1$ .

As a check for this analysis, we set  $s = 1$  in (4.4) and (4.5) to recover the solutions for surface wrinkling of a neo-Hookean film bonded to a

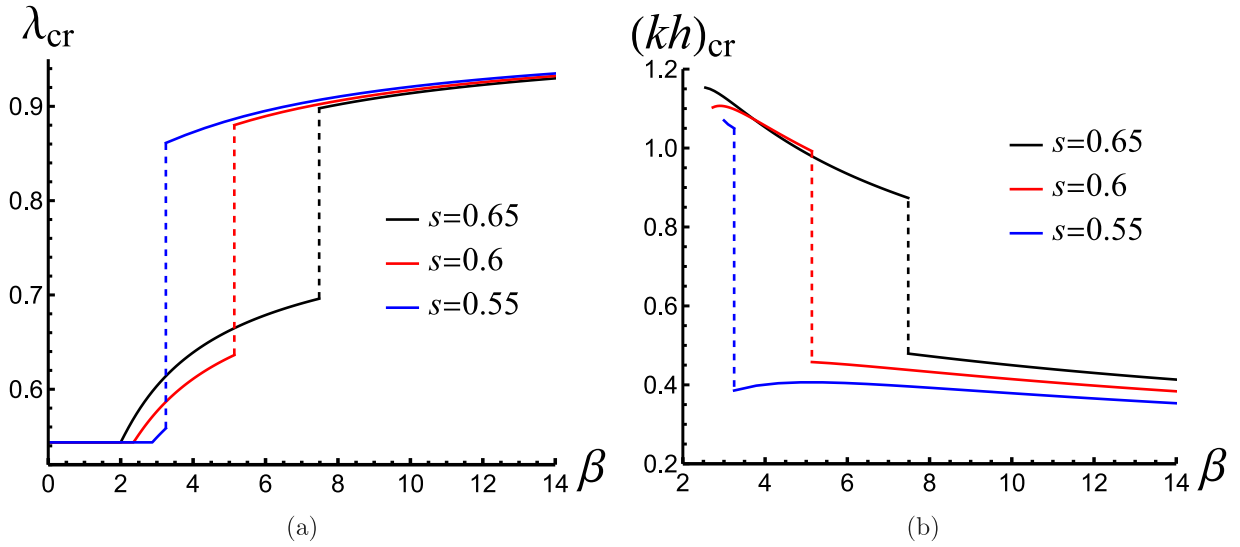


Fig. 8. The critical stretch  $\lambda_{cr}$  and the critical wavenumber  $(kh)_{cr}$  as functions of the modulus ratio  $\beta$  when  $\theta_0 = \pi/2$  and  $s < 1$ .

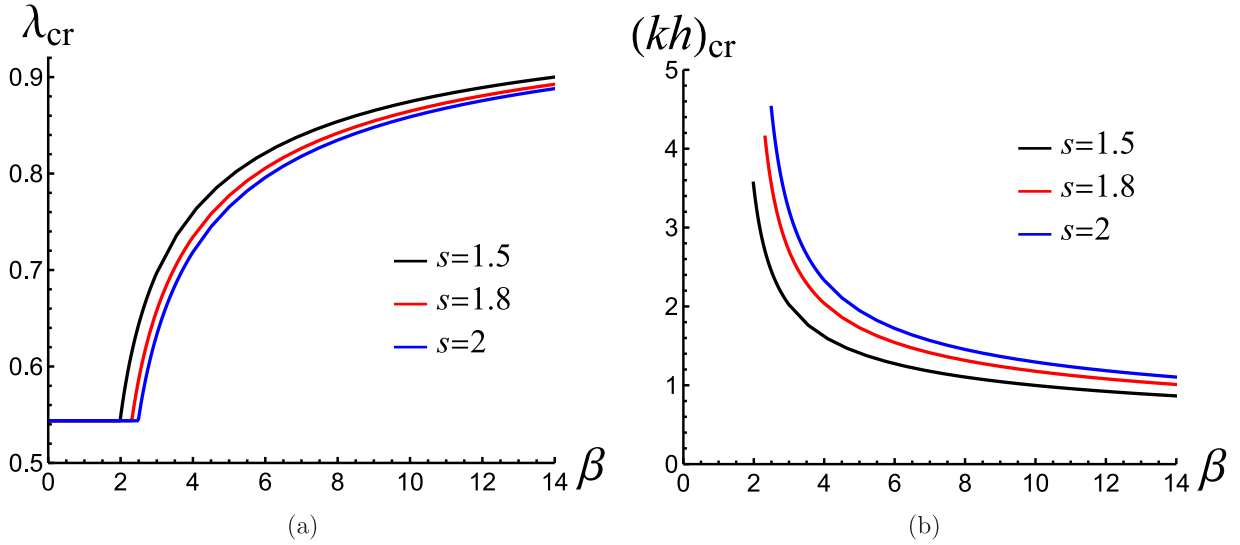


Fig. 9. The critical stretch  $\lambda_{cr}$  and the critical wavenumber  $(kh)_{cr}$  as functions of the modulus ratio  $\beta$  when  $\theta_0 = \pi/2$  and  $s > 1$ .

neo-Hookean substrate up to the truncated order (Cai and Fu, 1999; Alawiye et al., 2019; Wang et al., 2023).

In Fig. 11, a comparison between the exact solution and the asymptotic solution is given by specifying  $s = 0.8$ . A good agreement is found for both the critical stretch  $\lambda_{cr}$  and the critical wavenumber  $(kh)_{cr}$ , especially when  $\beta$  is large enough.

#### 4.2. The initial director is aligned in the vertical direction

Next, we consider the case  $\theta_0 = \pi/2$ . With a similar manner as before, we only consider  $s > 1$  and  $s \sim \mathcal{O}(1)$ . Applying the same solution procedure, we find  $\zeta_1$  and  $\zeta_2$  in (4.1):

$$\begin{aligned} \zeta_1 &= -\frac{6s^2 + \omega}{12\omega s}, \\ \zeta_2 &= \frac{180s^3\omega - 180s^2\omega - 900s^5 + 1440s^4 + 31s\omega^2 + 8\omega^2}{1440s^3\omega^2}. \end{aligned} \quad (4.6)$$

Inserting  $\omega = \beta(kh)^3$  back into (4.6), the asymptotic solution satisfying the bifurcation condition (3.8) is found:

$$\lambda = 1 - \frac{s}{2(kh)\beta} - \frac{(kh)^2}{12s} + \frac{(kh)^4(31s + 8)}{1440s^3} + \frac{s(8 - 5s)}{8(kh)^2\beta^2} + \frac{(s - 1)(kh)}{8s\beta} + \mathcal{O}((kh)^6). \quad (4.7)$$

Based on (3.11)<sub>2</sub> with  $i = 3$ , we obtain the asymptotic solutions:

$$\lambda_{cr} = 1 - \frac{3^{2/3}s^{1/3}}{4\beta^{2/3}} - \frac{3^{4/3}(s - 12)}{160s^{1/3}\beta^{4/3}} - \frac{3^{2/3}s^{1/3}}{8\beta^{5/3}} + \mathcal{O}(\beta^{-2}), \quad (4.8)$$

$$(kh)_{cr} = \frac{3^{1/3}s^{2/3}}{\beta^{1/3}} + \frac{(32s - 29)}{20\beta} + \mathcal{O}(\beta^{-5/3}). \quad (4.9)$$

It can be seen that both the anisotropy parameter  $s$  and the modulus ratio  $\beta$  appear at the leading-order terms. Unlike (4.4) and (4.5), the anisotropy parameter  $s$  is now in the numerator, but since  $s > 1$ , the effect on the bifurcation and the number of wrinkles is the same as the previous case.

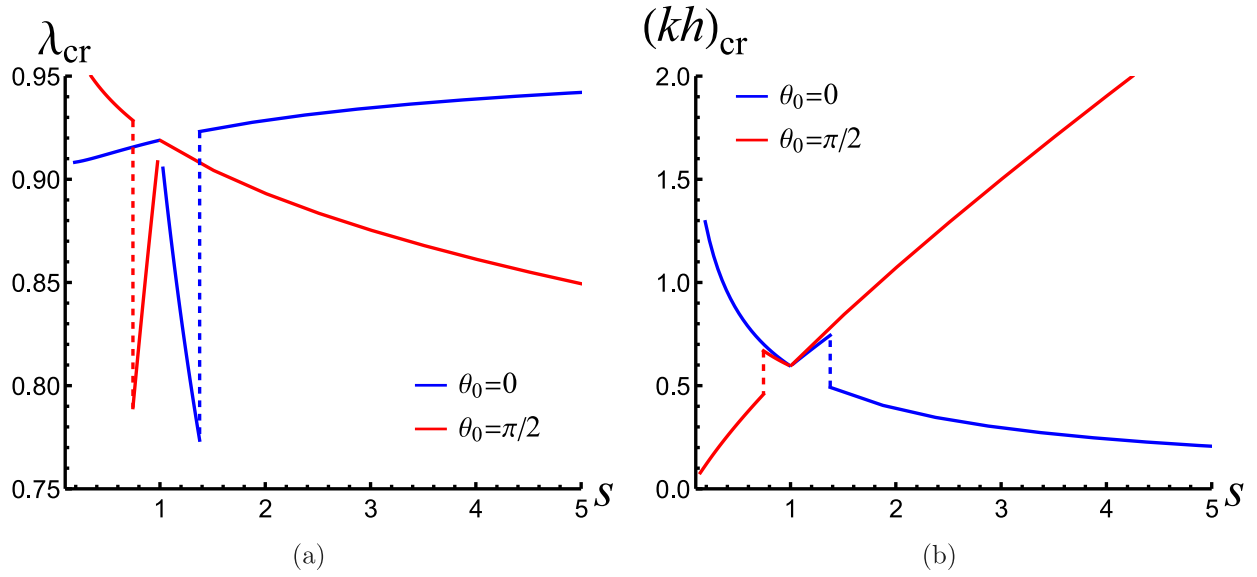


Fig. 10. The critical stretch  $\lambda_{cr}$  and the critical wavenumber  $(kh)_{cr}$  as functions of the anisotropy parameter  $s$  for when  $\beta = 15$  is fixed.

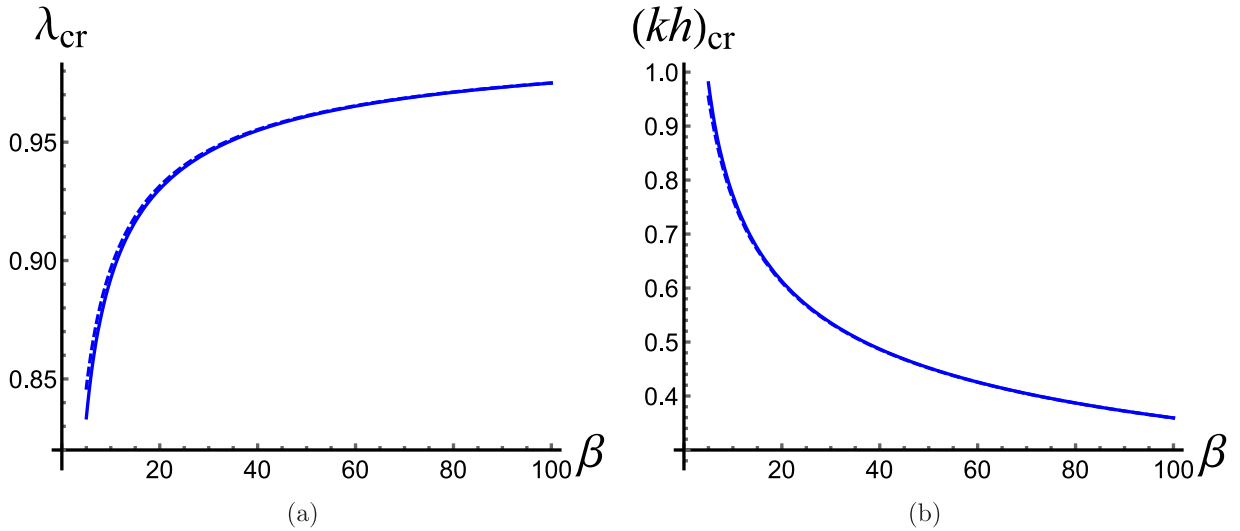


Fig. 11. Comparisons between the asymptotic solution (dashed line) and the exact solution (solid line) when  $\theta_0 = 0$  and  $s = 0.8$  where no director reorientation is allowed.

For  $s = 1$ , (4.8) and (4.9) reduce to the solutions for neo-Hookean bilayers (Cai and Fu, 1999; Alawiye et al., 2019; Wang et al., 2023). We illustrate a comparison between the exact solution and the asymptotic one in Fig. 12. Good agreement between them, especially for large  $\beta$ , is a further validation for our asymptotic solutions.

## 5. Conclusion

The compression-induced surface wrinkling of a neo-Hookean substrate coated by an LCE film was studied within the framework of finite elasticity. By adopting a neo-classical model of LCEs proposed by Warner et al. (1988), we characterized the homogeneous solutions generated by axial compression before surface wrinkling. In particular, we assumed that the initial director is aligned in either the horizontal or the vertical direction. In this case, the deformed director is along either the horizontal or vertical direction (Mihai and Goriely, 2021). To carry out a bifurcation analysis, we considered both an increment in the displacement and an increment in the director, which required augmenting the classical incremental equations for the displacement with

the incremental equations for the directors. An exact bifurcation condition for surface wrinkling was then obtained from which the critical stretch  $\lambda_{cr}$  as well as the critical wavenumber  $(kh)_{cr}$  were identified.

Our analysis revealed the effect of two key parameters, namely, the modulus ratio between the LCE film and the substrate  $\beta$  and the anisotropy parameter  $s$ , on the critical wrinkled state. For the modulus ratio  $\beta$ , it is well known that a larger  $\beta$  promotes surface wrinkling but reduces the number of wrinkles. However, with directors, the situation changes. If  $\theta_0 = 0$ , the director is initially in the  $E_1$ -direction, the critical stretch  $\lambda_{cr}$  increases as  $s$  increases without director reorientation. Moreover, the critical wavenumber  $(kh)_{cr}$  is a decreasing function of  $s$ . If  $\theta_0 = \pi/2$ , a reverse trend is observed for both  $\lambda_{cr}$  and  $(kh)_{cr}$ .

We point out that there are two effective ways to increase the stability of LCE film–substrate bilayer. The first is to trigger director reorientation if  $s$  falls into some targeted regions. In this way, the onset of surface wrinkling will be delayed and the associated wavenumber will be amplified, with more wrinkles created. Moreover, if  $s$  is out of the targeted region, i.e. director reorientation is not admitted, we shall let the major-axis of liquid crystal molecules coincide with the alignment of the director. Correspondingly, we select  $s > 1$  and  $\theta_0 =$

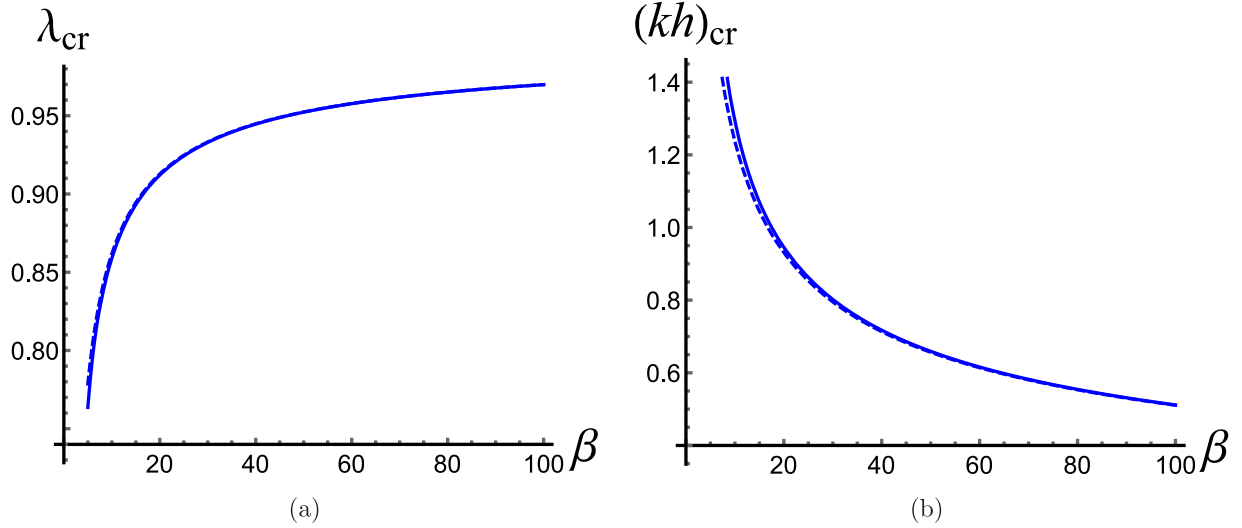


Fig. 12. Comparisons between the asymptotic solution (dashed line) and the exact solution (solid line) when  $\theta_0 = \pi/2$  and  $s = 2$  where no director reorientation is allowed.

$\pi/2$  or  $s < 1$  and  $\theta_0 = 0$ , then the bilayer is stabilized. Finally, we obtain some asymptotic solutions for  $\lambda_{cr}$  and  $(kh)_{cr}$  as  $kh \rightarrow 0$  and  $1/\beta \sim \mathcal{O}((kh)^3)$ , from which a clear picture on the combined effect of  $s$  and  $\beta$  emerges. Our work shows the importance of including the director in this type of instability and paves the way to a general incremental theory for LCEs.

It is further mentioned that surface wrinkling of LCE film–substrate bilayers can be complex due to a spacial distribution of the material parameter, a non-uniform distribution of initial directors, and three-dimensional geometry, etc. In spite of this, the current work aims to start from a simple setup and to further gain some fundamental knowledge of LCE wrinkling. For the issues of material inhomogeneity or stochastic properties, we refer to some recent papers (Mihai and Goriely, 2020a; Liu et al., 2024), and leave these for future studies.

#### CRedit authorship contribution statement

**Yang Liu:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Methodology, Investigation, Formal analysis, Conceptualization. **Qianqian Ji:** Writing – review & editing, Writing – original draft, Validation, Investigation, Formal analysis, Conceptualization. **Alain Goriely:** Writing – review & editing, Methodology, Formal analysis, Conceptualization.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

No data was used for the research described in the article.

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Grant No. EP/Y030559/1). Q.J. acknowledges the Tianjin Research Innovation Project, China for Postgraduate Students (Grant No. 2022SKY072). For the purpose of Open Access, the author has applied a CC BY public copyright license to any Author Accepted Manuscript (AAM) version arising from this submission. We thank Prof. Angela Mihai at Cardiff University for valuable discussions and comments on this work.

#### Appendix. The expressions of the coefficient matrices

The nonzero components of  $\mathbf{M}_1$  are given by

$$\begin{aligned}
 M_1^{11} &= e^{-\gamma_1 kh} (\gamma_1^2 + \lambda^4(s-1) + 1), & M_1^{12} &= e^{\gamma_1 kh} (\gamma_1^2 + \lambda^4(s-1) + 1), \\
 M_1^{13} &= e^{-\gamma_2 kh} (\gamma_2^2 + \lambda^4(s-1) + 1), & M_1^{14} &= e^{\gamma_2 kh} (\gamma_2^2 + \lambda^4(s-1) + 1), \\
 M_1^{21} &= \gamma_1 e^{-\gamma_1 kh} (\gamma_1^2 + \lambda^4(s-2) - 2), & M_1^{22} &= -\gamma_1 e^{\gamma_1 kh} (\gamma_1^2 + \lambda^4(s-2) - 2), \\
 M_1^{23} &= \gamma_2 e^{-\gamma_2 kh} (\gamma_2^2 + \lambda^4(s-2) - 2), & M_1^{24} &= -\gamma_2 e^{\gamma_2 kh} (\gamma_2^2 + \lambda^4(s-2) - 2), \\
 M_1^{31} &= -\gamma_1, & M_1^{32} &= \gamma_1, & M_1^{33} &= -\gamma_2, & M_1^{34} &= \gamma_2, & M_1^{35} &= -1, \\
 M_1^{36} &= -\lambda^2, \\
 M_1^{41} &= 1, & M_1^{42} &= 1, & M_1^{43} &= 1, & M_1^{44} &= 1, & M_1^{45} &= -1, & M_1^{46} &= -1, \\
 M_1^{51} &= \beta (\gamma_1^2 + \lambda^4(s-1) + 1), & M_1^{52} &= \beta (\gamma_1^2 + \lambda^4(s-1) + 1), \\
 M_1^{53} &= \beta (\gamma_2^2 + \lambda^4(s-1) + 1), & M_1^{54} &= \beta (\gamma_2^2 + \lambda^4(s-1) + 1), \\
 M_1^{55} &= -2, & M_1^{56} &= -\lambda^4 - 1, \\
 M_1^{61} &= \beta \gamma_1 (\gamma_1^2 + \lambda^4(s-2) - 2), & M_1^{62} &= -\beta \gamma_1 (\gamma_1^2 + \lambda^4(s-2) - 2), \\
 M_1^{63} &= \beta \gamma_2 (\gamma_2^2 + \lambda^4(s-2) - 2), & M_1^{64} &= -\beta \gamma_2 (\gamma_2^2 + \lambda^4(s-2) - 2), \\
 M_1^{65} &= -\lambda^4 - 1, & M_1^{66} &= -2\lambda^2.
 \end{aligned}$$

(A.1)

The nonzero components of  $\mathbf{M}_2$  are given by

$$\begin{aligned}
 M_2^{11} &= \frac{e^{-\gamma_3 kh} (\gamma_3^2 (\lambda^4 s^2 + s - 1) + \lambda^4 s^2)}{s^2}, \\
 M_2^{12} &= \frac{e^{\gamma_3 kh} (\gamma_3^2 (\lambda^4 s^2 + s - 1) + \lambda^4 s^2)}{s^2}, \\
 M_2^{13} &= \frac{e^{-\gamma_4 kh} (\gamma_4^2 (\lambda^4 s^2 + s - 1) + \lambda^4 s^2)}{s^2}, \\
 M_2^{14} &= \frac{e^{\gamma_4 kh} (\gamma_4^2 (\lambda^4 s^2 + s - 1) + \lambda^4 s^2)}{s^2},
 \end{aligned}$$

$$\begin{aligned}
M_2^{21} &= \frac{\gamma_3 e^{-\gamma_3 kh} (\gamma_3^2 (\lambda^4 s^2 + s - 1) + \lambda^4 s (-\lambda^4 s^2 + s - 3))}{s^2}, \\
M_2^{22} &= -\frac{\gamma_3 e^{\gamma_3 kh} (\gamma_3^2 (\lambda^4 s^2 + s - 1) + \lambda^4 s (-\lambda^4 s^2 + s - 3))}{s^2}, \\
M_2^{23} &= \frac{\gamma_4 e^{-\gamma_4 kh} (\gamma_4^2 (\lambda^4 s^2 + s - 1) + \lambda^4 s (-\lambda^4 s^2 + s - 3))}{s^2}, \\
M_2^{24} &= -\frac{\gamma_4 e^{\gamma_4 kh} (\gamma_4^2 (\lambda^4 s^2 + s - 1) + \lambda^4 s (-\lambda^4 s^2 + s - 3))}{s^2}, \\
M_2^{31} &= -\gamma_3, \quad M_2^{32} = \gamma_3, \quad M_2^{33} = -\gamma_4, \quad M_2^{34} = \gamma_4, \quad M_2^{35} = -1, \\
M_2^{36} &= -\lambda^2, \\
M_2^{41} &= 1, \quad M_2^{42} = 1, \quad M_2^{43} = 1, \quad M_2^{44} = 1, \quad M_2^{45} = -1, \quad M_2^{46} = -1, \\
M_2^{51} &= \beta \left( \lambda^4 + \frac{\gamma_3^2 (\lambda^4 s^2 + s - 1)}{s^2} \right), \\
M_2^{52} &= \beta \left( \lambda^4 + \frac{\gamma_3^2 (\lambda^4 s^2 + s - 1)}{s^2} \right), \\
M_2^{53} &= \beta \left( \lambda^4 + \frac{\gamma_4^2 (\lambda^4 s^2 + s - 1)}{s^2} \right), \\
M_2^{54} &= \beta \left( \lambda^4 + \frac{\gamma_4^2 (\lambda^4 s^2 + s - 1)}{s^2} \right), \\
M_2^{55} &= -2\lambda^4, \quad M_2^{56} = -\lambda^4 (\lambda^4 + 1), \\
M_2^{61} &= \frac{\beta \gamma_3 (\gamma_3^2 (\lambda^4 s^2 + s - 1) + \lambda^4 s (-\lambda^4 s^2 + s - 3))}{s^2}, \\
M_2^{62} &= \frac{\beta \gamma_3 (\lambda^4 s (\lambda^4 s^2 - s + 3) - \gamma_3^2 (\lambda^4 s^2 + s - 1))}{s^2}, \\
M_2^{63} &= \frac{\beta \gamma_4 (\gamma_4^2 (\lambda^4 s^2 + s - 1) + \lambda^4 s (-\lambda^4 s^2 + s - 3))}{s^2}, \\
M_2^{64} &= \frac{\beta \gamma_4 (\lambda^4 s (\lambda^4 s^2 - s + 3) - \gamma_4^2 (\lambda^4 s^2 + s - 1))}{s^2}, \\
M_2^{65} &= -\lambda^4 (\lambda^4 + 1), \quad M_2^{66} = -2\lambda^6.
\end{aligned}$$

(A.2)

The nonzero components of  $\mathbf{M}_3$  are given by

$$\begin{aligned}
M_3^{11} &= s e^{-\gamma_5 kh} (\lambda^4 + \gamma_5^2 (\lambda^4 + s - 1)), \\
M_3^{12} &= s e^{\gamma_5 kh} (\lambda^4 + \gamma_5^2 (\lambda^4 + s - 1)), \\
M_3^{13} &= s e^{-\gamma_6 kh} (\lambda^4 + \gamma_6^2 (\lambda^4 + s - 1)), \\
M_3^{14} &= s e^{\gamma_6 kh} (\lambda^4 + \gamma_6^2 (\lambda^4 + s - 1)), \\
M_3^{21} &= \gamma_5 e^{-\gamma_5 kh} (\gamma_5^2 s (\lambda^4 + s - 1) + \lambda^4 (-\lambda^4 + s - 3)), \\
M_3^{22} &= -\gamma_5 e^{\gamma_5 kh} (\gamma_5^2 s (\lambda^4 + s - 1) + \lambda^4 (-\lambda^4 + s - 3)), \\
M_3^{23} &= \gamma_4 e^{-\gamma_4 kh} (\gamma_4^2 s (\lambda^4 + s - 1) + \lambda^4 (-\lambda^4 + s - 3)), \\
M_3^{24} &= -\gamma_4 e^{\gamma_4 kh} (\gamma_4^2 s (\lambda^4 + s - 1) + \lambda^4 (-\lambda^4 + s - 3)), \\
M_3^{31} &= -\gamma_5, \quad M_3^{32} = \gamma_5, \quad M_3^{33} = -\gamma_6, \quad M_3^{34} = \gamma_6, \quad M_3^{35} = -1, \\
M_3^{36} &= -\lambda^2, \\
M_3^{41} &= 1, \quad M_3^{42} = 1, \quad M_3^{43} = 1, \quad M_3^{44} = 1, \quad M_3^{45} = -1, \quad M_3^{46} = -1, \\
M_3^{51} &= \beta s (\lambda^4 + \gamma_5^2 (\lambda^4 + s - 1)), \quad M_3^{52} = \beta s (\lambda^4 + \gamma_5^2 (\lambda^4 + s - 1)), \\
M_3^{53} &= \beta s (\lambda^4 + \gamma_6^2 (\lambda^4 + s - 1)), \quad M_3^{54} = \beta s (\lambda^4 + \gamma_6^2 (\lambda^4 + s - 1)), \\
M_3^{55} &= -2\lambda^4, \quad M_3^{56} = -\lambda^4 (\lambda^4 + 1), \\
M_3^{61} &= \beta \gamma_5 (\gamma_5^2 s (\lambda^4 + s - 1) + \lambda^4 (-\lambda^4 + s - 3)), \\
M_3^{62} &= \beta \gamma_5 (\lambda^4 (\lambda^4 - s + 3) - \gamma_5^2 s (\lambda^4 + s - 1)), \\
M_3^{63} &= \beta \gamma_6 (\gamma_6^2 s (\lambda^4 + s - 1) + \lambda^4 (-\lambda^4 + s - 3)), \\
M_3^{64} &= \beta \gamma_6 (\lambda^4 (\lambda^4 - s + 3) - \gamma_6^2 s (\lambda^4 + s - 1)), \\
M_3^{65} &= -\lambda^4 (\lambda^4 + 1), \quad M_3^{66} = -2\lambda^6,
\end{aligned}$$

(A.3)

where the eigenvalues  $\gamma_5$  and  $\gamma_6$  read

$$\begin{aligned}
\gamma_5 &= \left( \frac{\lambda^4 + 3 - 2s - \sqrt{(\lambda^4 + 3)^2 - 8(\lambda^4 + 1)s}}{2s(\lambda^4 + s - 1)} \right)^{1/2} \lambda^2, \\
\gamma_6 &= \left( \frac{\lambda^4 + 3 - 2s + \sqrt{(\lambda^4 + 3)^2 - 8(\lambda^4 + 1)s}}{2s(\lambda^4 + s - 1)} \right)^{1/2} \lambda^2.
\end{aligned}$$

The nonzero components of  $\mathbf{M}_4$  are given by

$$\begin{aligned}
M_4^{11} &= \frac{e^{-\gamma_7 kh} (-\lambda^4 + (\gamma_7^2 + 1)s^2 + \lambda^4 s)}{s}, \\
M_4^{12} &= \frac{e^{\gamma_7 kh} (-\lambda^4 + (\gamma_7^2 + 1)s^2 + \lambda^4 s)}{s}, \\
M_4^{13} &= \frac{e^{-\gamma_8 kh} (-\lambda^4 + (\gamma_8^2 + 1)s^2 + \lambda^4 s)}{s}, \\
M_4^{14} &= \frac{e^{\gamma_8 kh} (-\lambda^4 + (\gamma_8^2 + 1)s^2 + \lambda^4 s)}{s}, \\
M_4^{21} &= \frac{\gamma_7 e^{-\gamma_7 kh} (-2\lambda^4 + (\gamma_7^2 - 2)s^2 + \lambda^4 s)}{s}, \\
M_4^{22} &= -\frac{\gamma_7 e^{\gamma_7 kh} (-2\lambda^4 + (\gamma_7^2 - 2)s^2 + \lambda^4 s)}{s}, \\
M_4^{23} &= \frac{\gamma_8 e^{-\gamma_8 kh} (-2\lambda^4 + (\gamma_8^2 - 2)s^2 + \lambda^4 s)}{s}, \\
M_4^{24} &= -\frac{\gamma_8 e^{\gamma_8 kh} (-2\lambda^4 + (\gamma_8^2 - 2)s^2 + \lambda^4 s)}{s}, \\
M_4^{31} &= -\gamma_7, \quad M_4^{32} = \gamma_7, \quad M_4^{33} = -\gamma_8, \quad M_4^{34} = \gamma_8, \quad M_4^{35} = -1, \\
M_4^{36} &= -\lambda^2, \\
M_4^{41} &= 1, \quad M_4^{42} = 1, \quad M_4^{43} = 1, \quad M_4^{44} = 1, \quad M_4^{45} = -1, \quad M_4^{46} = -1, \\
M_4^{51} &= \beta \left( \lambda^4 + \gamma_7^2 s - \frac{\lambda^4}{s} + s \right), \quad M_4^{52} = \beta \left( \lambda^4 + \gamma_7^2 s - \frac{\lambda^4}{s} + s \right), \\
M_4^{53} &= \beta \left( \lambda^4 + \gamma_8^2 s - \frac{\lambda^4}{s} + s \right), \quad M_4^{54} = \beta \left( \lambda^4 + \gamma_8^2 s - \frac{\lambda^4}{s} + s \right), \\
M_4^{55} &= -2, \quad M_4^{56} = -\lambda^4 - 1, \\
M_4^{61} &= \frac{\beta \gamma_7 (-2\lambda^4 + (\gamma_7^2 - 2)s^2 + \lambda^4 s)}{s}, \\
M_4^{62} &= -\frac{\beta \gamma_7 (-2\lambda^4 + (\gamma_7^2 - 2)s^2 + \lambda^4 s)}{s}, \\
M_4^{63} &= \frac{\beta \gamma_8 (-2\lambda^4 + (\gamma_8^2 - 2)s^2 + \lambda^4 s)}{s}, \\
M_4^{64} &= -\frac{\beta \gamma_8 (-2\lambda^4 + (\gamma_8^2 - 2)s^2 + \lambda^4 s)}{s}, \\
M_4^{65} &= -\lambda^4 - 1, \quad M_4^{66} = -2\lambda^2,
\end{aligned}$$

(A.4)

where the eigenvalues  $\gamma_7$  and  $\gamma_8$  read

$$\begin{aligned}
\gamma_7 &= \left( \frac{3\lambda^4 + s^2 - 2\lambda^4 s - \sqrt{s^4 + 2\lambda^4 s^2 (3 - 4s) + \lambda^8 (9 - 8s)}}{2s^2} \right)^{1/2}, \\
\gamma_8 &= \left( \frac{3\lambda^4 + s^2 - 2\lambda^4 s + \sqrt{s^4 + 2\lambda^4 s^2 (3 - 4s) + \lambda^8 (9 - 8s)}}{2s^2} \right)^{1/2}.
\end{aligned}$$

## References

- Agrawal, A., Luchette, P., Palfy-Muhoray, P., Biswal, S.L., Chapman, W.G., Verduzco, R., 2012. Surface wrinkling in liquid crystal elastomers. *Soft Matter* 8 (27), 7138–7142.
- Agrawal, A., Yun, T., Pesek, S.L., Chapman, W.G., Verduzco, R., 2014. Shape-responsive liquid crystal elastomer bilayers. *Soft Matter* 10 (9), 1411–1415.
- Alawiye, H., Kuhl, E., Goriely, A., 2019. Revisiting the wrinkling of elastic bilayers I: linear analysis. *Phil. Trans. R. Soc. A* 377, 20180076.

- Anderson, D.R., Carlson, D.E., Fried, E., 1999. A continuum-mechanical theory for nematic elastomers. *J. Elasticity* 56 (1), 33–58.
- Andres, S., Steinmann, P., Budday, S., 2018. The origin of compression influences geometric instabilities in bilayers. *Proc. R. Soc. A Math. Phys. Eng. Sci.* 474 (2217), 20180267.
- Ball, J.M., Bedford, S.J., 2015. Discontinuous order parameters in liquid crystal theories. *Mol. Cryst. Liq. Cryst.* 612 (1), 1–23.
- Barnes, M., Feng, F., Biggins, J., 2023. Surface instability in a nematic elastomer. *Phys. Rev. Lett.* 131, 238101.
- Biot, M.A., 1963. Surface instability of rubber in compression. *Appl. Sci. Res.* 12, 168–182.
- Biot, M.A., 1965. *Mechanics of Incremental Deformations*. John Wiley & Sons, New York.
- Bladon, P., Terentjev, E.M., Warner, M., 1993. Transitions and instabilities in liquid crystal elastomers. *Phys. Rev. E* 47 (6), R3838–R3840.
- Budday, S., Kuhl, E., Hutchinson, J.W., 2015. Period-doubling and period-tripling in growing bilayered systems. *Phil. Mag.* 95, 3208–3224.
- Cai, Z., Fu, Y.B., 1999. On the imperfection sensitivity of a coated elastic half-space. *Proc. R. Soc. Lond. Ser. A Math. Phys. Eng. Sci.* 455 (1989), 3285–3309.
- Cao, Y.P., Hutchinson, J.W., 2012. Wrinkling phenomena in neo-Hookean film/substrate bilayers. *J. Appl. Mech.-Trans. ASME* 79 (3), 031019.
- Chen, Y.C., Fried, E., 2006. Uniaxial nematic elastomers: constitutive framework and a simple application. *Proc. R. Soc. Lond. Ser. A Math. Phys. Eng. Sci.* 462, 20140494.
- Chen, M., Hou, Y., An, R., Qi, H.J., Zhou, K., 2023. 4D printing of reprogrammable liquid crystal elastomers with synergistic photochromism and photoactuation. *Adv. Mater.* <http://dx.doi.org/10.1002/adma.202303969>, (in press).
- Chen, Y., Kuentzler, A.S., Hayward, R.C., Jin, L., 2022. Formation of rolls from liquid crystal elastomer bistraps. *Soft Matter* 18 (21), 4077–4089.
- Conti, S., DeSimone, A., Dolzmann, G., 2002. Soft elastic response of stretched sheets of nematic elastomers: a numerical study. *J. Mech. Phys. Solids* 50 (7), 1431–1451.
- de Gennes, P.G., Hébert, M., Kant, R., 1997. Artificial muscles based on nematic gels. In: *Macromolecular Symposia*, Vol. 113, No. 1. pp. 39–49.
- De Pascalis, R., Destrade, M., Goriely, A., 2011. Nonlinear correction to the Euler buckling formula for compressed cylinders with guided-guided end conditions. *J. Elasticity* 102, 191–200.
- DeSimone, A., Gidoni, P., Noselli, G., 2015. Liquid crystal elastomer strips as soft crawlers. *J. Mech. Phys. Solids* 84, 254–272.
- Fried, E., Korchagin, V., 2002. Striping of nematic elastomer. *Int. J. Solids Struct.* 39, 3451–3467.
- Fried, E., Sellers, S., 2006. Soft elasticity is not necessary for striping in nematic elastomers. *J. Appl. Phys.* 100 (4), 043521.
- Fu, Y.B., Jin, L., Goriely, A., 2021. Necking, beading, and bulging in soft elastic cylinders. *J. Mech. Phys. Solids* 147, 104250.
- Fu, Y.B., Ogden, R.W., 1999. Nonlinear stability analysis of pre-stressed elastic bodies. *Contin. Mech. Thermodyn.* 11 (3), 141–172.
- Fu, Y.X., Wang, Y.S., Fu, Y.B., 2023. New mountain ridge modes in a film/substrate bilayer. *Math. Mech. Solids* 1–15. <http://dx.doi.org/10.1177/10812865231208414>.
- Fu, C., Xu, F., Huo, Y., 2018. Photo-controlled patterned wrinkling of liquid crystalline polymer films on compliant substrates. *Int. J. Solids Struct.* 132, 264–277.
- Goriely, A., 2017. *The Mathematics and Mechanics of Biological Growth*. Springer, New York.
- Goriely, A., Mihai, L.A., 2021. Liquid crystal elastomers wrinkling. *Nonlinearity* 34 (8), 5599–5629.
- Goriely, A., Moulton, D.E., Mihai, L.A., 2023. A rod theory for liquid crystalline elastomers. *J. Elasticity* 153, 509–532.
- Goriely, A., Vandiver, R., Destrade, 2008. *Proc. R. Soc. A Math. Phys. Eng. Sci.* 464, 3003–3019.
- Holland, M., Budday, S., Goriely, A., Kuhl, E., 2018. Symmetry breaking in wrinkling patterns: Gyri are universally thicker than sulci. *Phys. Rev. Lett.* 121, 228002.
- Jia, F., Pearce, S.P., Goriely, A., 2018. Curvature delays growth-induced wrinkling. *Phys. Rev. E* 98, 033003.
- Jin, L., Liu, Y., Cai, Z., 2019. Post-buckling analysis on growing tubular tissues: A semi-analytical approach and imperfection sensitivity. *Int. J. Solids Struct.* 162, 121–134.
- Krieger, M.S., Dias, M.A., 2019. Tunable wrinkling of thin nematic liquid crystal elastomer sheets. *Phys. Rev. E* 100 (2), 022701.
- Kundler, I., Finkelmann, H., 1995. Strain-induced director reorientation in nematic liquid single crystal elastomers. *Macromol. Rapid Commun.* 16 (9), 679–686.
- Lee, V., Wihardja, A., Bhattacharya, K., 2023. A macroscopic constitutive relation for isotropic-genesis, polydomain liquid crystal elastomers. *J. Mech. Phys. Solids* 179, 105369.
- Li, B., Cao, Y.P., Feng, X.Q., Gao, H., 2012. Mechanics of morphological instabilities and surface wrinkling in soft materials: a review. *Soft Matter* 8 (5728).
- Liu, Y., 2023. Higher order solution to the Euler buckling threshold for compressible hyperelastic bilayers. *Acta Mech. Sinica* 39 (8), 422379.
- Liu, Y., Dai, H.-H., 2014. Compression of a hyperelastic layer-substrate structure: Transitions between buckling and surface modes. *Internat. J. Engrg. Sci.* 80, 74–89.
- Liu, Y., Dorfmann, L., 2023. Localized necking and bulging of finitely deformed residually stressed solid cylinder. *Math. Mech. Solids* 1–23. <http://dx.doi.org/10.1177/10812865231186951>.
- Liu, S., Huang, K., Wang, K., Wang, B., 2022. Programmable deformation of liquid crystal elastomer plates subjected to concentrated light illumination. *Mech. Mater.* 175, 104501.
- Liu, R.C., Liu, Y., Cai, Z., 2021a. Influence of the growth gradient on surface wrinkling and pattern transition in growing tubular tissues. *Proc. R. Soc. A Math. Phys. Eng. Sci.* 477, 20210441.
- Liu, R.C., Liu, Y., Goriely, A., 2024. Surface wrinkling of a film coated to a graded substrate. *J. Mech. Phys. Solids* 186, 105603.
- Liu, Y., Ma, W., Dai, H.-H., 2020a. On a consistent finite-strain plate model of nematic liquid crystal elastomers. *J. Mech. Phys. Solids* 145, 104169.
- Liu, Y., Ma, W., Dai, H.-H., 2021b. Bending-induced director reorientation of a nematic liquid crystal elastomer bonded to a hyperelastic substrate. *J. Appl. Phys.* 129 (10), 104701.
- Liu, H., Tian, H., Shao, J., Wang, Z., Li, X., Wang, C., Chen, X., 2020b. An electrically actuated soft artificial muscle based on a high-performance flexible electrothermal film and liquid-crystal elastomer. *ACS Appl. Mater. Interfaces* 12 (50), 56338–56349.
- Mahardika, N., Raistrick, T., Mihai, L.A., Wang, H., 2024. All-atom molecular dynamics simulations of nematic liquid crystal elastomers. *Int. J. Solids Struct.* 291, 112717.
- Mbanga, B.L., Ye, F., Selinger, J.V., Selinger, R.L., 2010. Modeling elastic instabilities in nematic elastomers. *Phys. Rev. E* 82 (5), 051701.
- Mihai, L.A., Gagliardi, A., Terentjev, E., Goriely, A., 2023a. Anti-hertz bulging of actuated liquid crystal elastomers. *Extreme Mech. Lett.* 64, 102066.
- Mihai, L.A., Goriely, A., 2020a. Likely striping in stochastic nematic elastomers. *Math. Mech. Solids* 25 (10), 1851–1872.
- Mihai, L.A., Goriely, A., 2020b. A plate theory for nematic liquid crystalline solids. *J. Mech. Phys. Solids* 144, 104101.
- Mihai, L.A., Goriely, A., 2021. Instabilities in liquid crystal elastomers. *MRS Bull.* 46, 784–794.
- Mihai, L.A., Mistry, D., Raistrick, T., Gleeson, H.F., Goriely, A., 2022. A mathematical model for the auxetic response of liquid crystal elastomers. *Phil. Trans. R. Soc. A* 380, 20210326.
- Mitchell, G.R., Davis, F., Guo, W., 1993. Strain-induced transitions in liquid-crystal elastomers. *Phys. Rev. Lett.* 71, 2947–2950.
- Ogden, R.W., 1984. *Non-Linear Elastic Deformations*. Ellis Horwood, New York.
- Pang, X., Lv, J.A., Zhu, C., Qin, L., Yu, Y., 2019. Photodeformable azobenzene-containing liquid crystal polymers and soft actuators. *Adv. Mater.* 31 (52), 1904224.
- Peng, Z., Jiang, Y., Chen, Y., Huo, Y., 2023. Attenuating liquid crystal elastomers' stress concentration by programming initial orientation. *Int. J. Mech. Sci.* 249, 108274.
- Potekhin, A., Wang, C., 2021. Numerical simulation and experimental validation of bending and curling behaviors of liquid crystal elastomer beams under thermal actuation. *Appl. Phys. Lett.* 118, 241903.
- Qing, X., Liu, Y., Wei, J., Zheng, R., Zhu, C., Yu, Y., 2019. Phototunable morpho butterfly microstructures modified by liquid crystal polymers. *Adv. Opt. Mater.* 7 (3), 1801494.
- Soni, H., Pelcovits, R.A., Powers, T.R., 2016. Wrinkling of a thin film on a nematic liquid-crystal elastomer. *Phys. Rev. E* 94 (1), 012701.
- Sonnet, A.M., Virga, E.G., 2022. Model for a photoresponsive nematic elastomer ribbon. *J. Elasticity* 1–28.
- Stewart, P.S., Waters, S.L., El Sayed, T., Vella, D., Goriely, A., 2016. Wrinkling, creasing, and folding in fiber-reinforced soft tissues. *Extreme Mech. Lett.* 8, 22–29.
- Tian, H., Wang, Z., Chen, Y., Shao, J., Gao, T., Cai, S., 2018. Polydopamine-coated main-chain liquid crystal elastomer as optically driven artificial muscle. *ACS Appl. Mater. Interfaces* 10 (9), 8307–8316.
- Urayama, K., Honda, S., Takigawa, T., 2006. Deformation coupled to director rotation in swollen nematic elastomers under electric fields. *Macromolecules* 39 (5), 1943–1949.
- Urayama, K., Kondo, H., Arai, Y.O., Takigawa, T., 2005. Electrically driven deformations of nematic gels. *Phys. Rev. E* 71 (5), 051713.
- Wang, Y., An, J., Lee, H., 2022a. Recent advances in molecular programming of liquid crystal elastomers with additive manufacturing for 4D printing. *Mol. Syst. Des. Eng.* 7 (12), 1588–1601.
- Wang, Z., Chehade, A.E.H., Govindjee, S., Nguyen, T.D., 2022c. A nonlinear viscoelasticity theory for nematic liquid crystal elastomers. *J. Mech. Phys. Solids* 163, 104829.
- Wang, M., Han, Y., Guo, L.X., Lin, B.P., Yang, H., 2019a. Photocontrol of helix handedness in curled liquid crystal elastomers. *Liq. Cryst.* 46 (8), 1231–1240.
- Wang, Z., He, Q., Wang, Y., Cai, S., 2019b. Programmable actuation of liquid crystal elastomers via living exchange reaction. *Soft Matter* 15 (13), 2811–2816.
- Wang, Y., Huang, X., Zhang, J., Bi, M., Zhang, J., Niu, H., Jiang, H., 2017a. Two-step crosslinked liquid-crystalline elastomer with reversible two-way shape memory characteristics. *Mol. Cryst. Liq. Cryst.* 650 (1), 13–22.
- Wang, G., Liu, Y., Fu, Y.B., 2023. A refined model for the buckling of film/substrate bilayers. *Math. Mech. Solids* 28 (1), 313–330.
- Wang, Z., Tian, H., He, Q., Cai, S., 2017b. Reprogrammable, reprocessable, and self-healable liquid crystal elastomer with exchangeable disulfide bonds. *ACS Appl. Mater. Interfaces* 9 (38), 33119–33128.
- Ware, T.H., Biggins, J.S., Shick, A.F., Mark Warner, M., White, T.J., 2016. Localized soft elasticity in liquid crystal elastomers. *Nature Commun.* 7 (10781).

- Warner, M., Gelling, K.P., Vilgis, T.A., 1988. Theory of nematic networks. *J. Chem. Phys.* 88, 4008–4013.
- Warner, M., Terentjev, E.M., 2007. *Liquid Crystal Elastomers*. Oxford University Press.
- White, T.J., Broer, D.J., 2015. Programmable and adaptive mechanics with liquid crystal polymer networks and elastomers. *Nature Mater.* 14, 1087–1098.
- Wu, J., Xiao, Y., Zhang, Y., Fang, D., 2023. A constitutive model of liquid crystal elastomers with loading-history dependence. *J. Mech. Phys. Solids* 174, 105258.
- Xiao, Y., Wu, J., Zhang, Y., 2023. Recent advances in the design, fabrication, actuation mechanisms and applications of liquid crystal elastomers. *Soft Sci.* 3, 11.
- Xu, Y., Zhang, Y., Huo, Y., 2020. Electric-field induced deformation and bending in nematic elastomer strips with orientation gradient. *Int. J. Solids Struct.* 202, 243–259.
- Xu, F., Zhao, S., 2020. Thermal wrinkling of liquid crystal polymer shell/core spheres. *Extreme Mech. Lett.* 40, 100860.
- Yan, Z., Ji, X., Wu, W., Wei, J., Yu, Y., 2012. Light-switchable behavior of a microarray of azobenzene liquid crystal polymer induced by photodeformation. *Macromol. Rapid Commun.* 33 (16), 1362–1367.
- Zeng, H., Wani, O.M., Wasylczyk, P., Primagi, A., 2018. Light-driven, caterpillar-inspired miniature inching robot. *Macromol. Rapid Commun.* 39 (1), 1700224.
- Zhai, F., Feng, Y., Li, Z., Xie, Y., Ge, J., Wang, H., Qiu, W., Feng, W., 2021. 4D-printed untethered self-propelling soft robot with tactile perception: Rolling, racing, and exploring. *Matter* 4 (10), 3313–3326.
- Zhang, Y., Xuan, C., Jiang, Y., Huo, Y., 2019. Continuum mechanical modeling of liquid crystal elastomers as dissipative ordered solids. *J. Mech. Phys. Solids* 126, 285–303.
- Zhao, S., Chen, Y., Huo, Y., 2023. Formation of lamellar domains in liquid crystal elastomers under compression. *Int. J. Mech. Sci.* 247, 108185.
- Zhao, S., Fu, C.Xu.F., Huo, Y., 2019. Controllable wrinkling patterns on liquid crystal polymer film/substrate systems by laser illumination. *Extreme Mech. Lett.* 30, 100502.
- Zhao, T., Wang, J., Fan, Y., Dou, W., 2022. Helical liquid crystal elastomer miniature robot with photocontrolled locomotion. *Adv. Mater. Technol.* 7 (10), 2200222.