



## Electro-mechanical wrinkling of soft dielectric films bonded to hyperelastic substrates

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### ABSTRACT

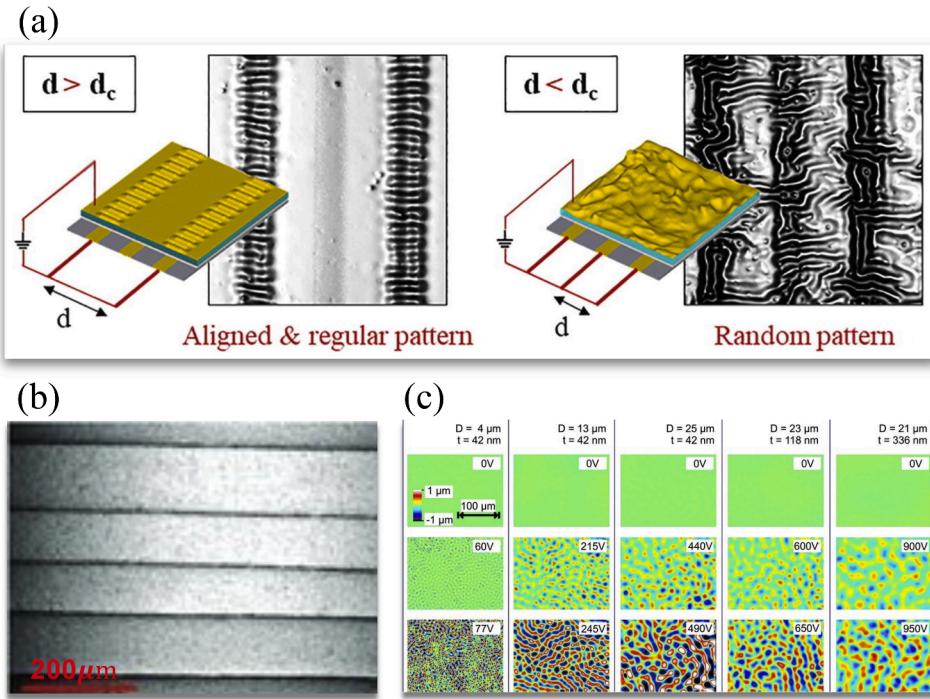
Active control of wrinkling in soft film-substrate composites using electric fields is a critical challenge in tunable material systems. Here, we investigate the electro-mechanical instability of a soft dielectric film bonded to a hyperelastic substrate, revealing the fundamental mechanisms that enable on-demand surface patterning. For the linearized stability analysis, we use the Stroh formalism and the surface impedance method to obtain exact and sixth-order approximate bifurcation equations that signal the onset of wrinkles. We derive the explicit bifurcation equations giving the critical stretch and critical voltage for wrinkling, as well as the corresponding critical wavenumber. We look at scenarios where the voltage is kept constant and the stretch changes, and vice versa. We provide the thresholds of the shear modulus ratio  $r_c^0$  or pre-stretch  $\lambda_c^0$  below which the film-substrate system wrinkles mechanically, prior to the application of a voltage. These predictions offer theoretical guidance for practical structural design, as the shear modulus ratio  $r$  and/or the pre-stretch  $\lambda$  can be chosen to be slightly greater than  $r_c^0$  and/or  $\lambda_c^0$ , so that the film-substrate system wrinkles with a small applied voltage. Finally, we simulate the full nonlinear behavior using the Finite Element method (FEniCS) to validate our formulas and conduct a post-buckling analysis. This work advances the fundamental understanding of electro-mechanical wrinkling instabilities in soft material systems. By enabling active control of surface morphologies via applied electric fields, our findings open new avenues for adaptive technologies in soft robotics, flexible electronics, smart surfaces, and bioinspired systems.

### 1. Introduction

Surface wrinkling of soft materials and biological tissues is ubiquitous in nature and engineering (Li et al., 2012; Tan et al., 2020), typically occurring when a soft substrate coated with a stiffer film is loaded mechanically beyond a critical threshold (Liu et al., 2024). In biology, countless wrinkling morphologies appear, such as the folds of brain matter (Griffiths et al., 2009; Fernández et al., 2016; Balbi et al., 2020; Riccobelli and Bevilacqua, 2020), the track of oesophageal mucosa (Li et al., 2011), and the wrinkles of skin

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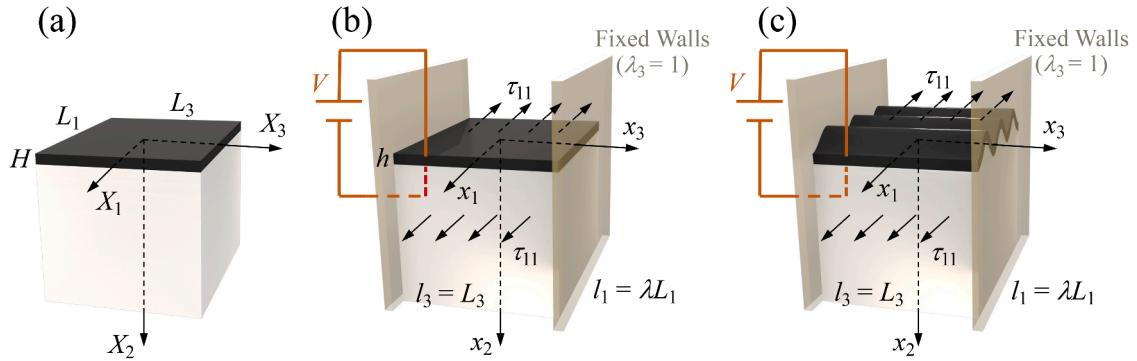
**Fig. 1.** Examples of electrically induced surface instabilities in soft polymer films. These patterns are formed not on a soft substrate as studied in this work, but on a rigid one, which limits the system's deformability but still illustrates key principles of electro-responsive wrinkling. (a) Wrinkle patterns can be switched from regular to random by tuning the spacing  $d$  between the underlying electrodes (Lin et al., 2020); (b) Highly aligned parallel lines can be formed by applying a voltage to a uniaxially pre-stretched film (Wang et al., 2012); (c) On films without pre-stretch, increasing the voltage causes a flat surface to buckle into random wrinkles and craters above a critical threshold (Ende et al., 2013).

(Autumn et al., 2002; Zhao et al., 2020b; Wang et al., 2024). In engineering, the wrinkling of film-substrate systems can be harnessed to design specific patterns and alter optical properties (Li et al., 2017), probe the surface characteristics of materials (Stafford et al., 2004), reduce effective surface tension (Lee et al., 2021), design novel nonlithographic phase masks (Zhao et al., 2020a), and help design novel flexible sensors (Wang et al., 2022; Lee et al., 2022; Yin et al., 2024), etc. Therefore, exploring the buckling and post-buckling regimes of film-substrate systems helps us understand and further control multiple pattern formations (Lai et al., 2025). However, the purely mechanical actuation of wrinkling/creasing in film-substrate systems does not allow for efficient active control of such surface patterns (Psarra et al., 2017).

The emergence of soft smart materials provides a great opportunity for applied research on film-substrate systems. Specifically, soft dielectric elastomers (DEs), which deform significantly under an external electric field (Pelrine et al., 1998; Guo et al., 2021), offer the advantages of extensive actuation strains (Li et al., 2013), fast response (Chen et al., 2019), and low elastic modulus (Shian et al., 2015), enabling their use in artificial muscles (Brochu and Pei, 2009), electrical energy storage devices (Li et al., 2018), sensors (Pelrine et al., 1998; Lee et al., 2022; Yin et al., 2024), grippers (Shian et al., 2015), and soft robots (Liang et al., 2020; Guo et al., 2021).

The coupling of Maxwell's equations of electricity with those of continuum mechanics complicates the study of film-substrate instabilities. For pure elastic film-substrate systems, early studies concentrated on analyzing linearized stability in the neighborhood of large contractions (Shield et al., 1994; Ogden and Sotiropoulos, 1996; Cai and Fu, 1999; Liu, 2023), and were followed by advanced explorations of wrinkling, post-buckling, semi-analytic methods, and finite element simulations (Cai and Fu, 2000; Cao and Hutchinson, 2011, 2012; Hutchinson, 2013; Fu and Ciarletta, 2015; Cheewaruangroj and Biggins, 2019; Alawiye et al., 2019, 2020; Liu et al., 2024). Regarding pure DE systems, many works have focused on instability (Bertoldi and Gei, 2011; Fu et al., 2018; Su et al., 2018, 2019b, 2023, 2024; Liguori and Gei, 2023; Zhu et al., 2024; Si et al., 2025).

Kofod et al. (2003), Wang et al. (2011a,b) and Wang and Zhao (2013) carried out a series of experiments on pre-stretched elastic dielectrics bonded to rigid substrates and subjected to high voltages, leading to localized creasing-like instabilities (see Fig. 1), which were later studied theoretically and numerically by Hutchinson (2021), Landis et al. (2022), and Li et al. (2024). In those cases, however, the rigidity of the substrates imposes significant limitations on applications. Systems comprising a dielectric film bonded to a soft substrate have also been studied: for example, Su et al. (2020b) investigated the bending deformation of a dielectric-elastic bilayer in response to a voltage; Almamo et al. (2024) studied the axisymmetric vibrations of a dielectric-elastic tubular bilayer system; and Sriram et al. (2024) used a data-driven approach to model the onset of wrinkling in composite DE bilayer structures subjected to combined electro-mechanical loading conditions.



**Fig. 2.** Schematic diagram of a soft dielectric film bonded to a hyperelastic substrate, confined between two lubricated rigid walls. (a) Initial undeformed configuration; (b) Current deformed configuration, prior to (c) the onset of wrinkles.

The conclusion of this survey is that the potential instabilities and pattern formation of DE films bonded to a soft hyperelastic substrate (Fig. 2) are yet to be analyzed theoretically and numerically. This work combines the advantages of DEs (a type of smart material) and film-substrate systems to investigate the stability of a soft dielectric film bonded to a hyperelastic substrate under a plane-strain mechanical load and a uniform transverse electric field (or voltage) (Fig. 2(b)). We work within the framework of nonlinear electro-elasticity theory and the associated linearized incremental theory developed by Verma and Chaudhury (1966) and Dorfmann and Ogden (2014). To overcome the complexity of conventional displacement-based methods, we use the Stroh formulation and the surface impedance method (Su et al., 2018) to derive exact solutions and approximate explicit expressions for the bifurcation equations. In addition, we use the finite element method based on FEniCS to conduct a wrinkling analysis of the DE film-substrate system, and the results are compared with the theoretical solutions. Finally, a post-buckling analysis of the DE film-substrate system is also conducted.

Our results show that the onset of wrinkling in a soft dielectric film-substrate system can be actively tuned by electro-mechanical loading, provided the material parameters are chosen appropriately. In particular, we find explicit formulas for the critical stretch and critical voltage at which wrinkles emerge, along with the corresponding wrinkle wavelength.

We look at two loading path scenarios: first, the applied voltage is fixed at the equilibrium value at which there is no applied traction, and a mechanical load is applied; second, the stretch is fixed at a given contractile or extensile level, and the voltage is increased. The analytical bifurcation results reveal a threshold stiffness ratio and pre-stretch (denoted  $r_c^0$  and  $\lambda_c^0$ ) below which the film-substrate system wrinkles under purely mechanical compression, even with no voltage applied. Unsurprisingly, this corresponds to the bifurcation criterion for the compression of non-coupled, hyperelastic systems. Above these thresholds, however, the film remains flat until a sufficient voltage triggers the instability, in the contractile as well as the extensile regimes. This behavior provides a practical design guideline: by selecting the substrate-to-film stiffness ratio and pre-stretch just above  $r_c^0$  and  $\lambda_c^0$ , one can ensure the system stays smooth under mechanical load and then wrinkles *on demand* with a small applied voltage (note that if the system is pre-stretched in extension, a potentially high voltage is required for wrinkling). We verify these predictions through finite element simulations, which not only confirm the accuracy of the critical stretch and voltage estimates, but also capture the post-buckling evolution of the wrinkle patterns, including the potential development of period-doubling and period-tripling patterns. Importantly, our stability analysis indicates that the wrinkle formation is a supercritical bifurcation in most cases, meaning the pattern amplitude grows gradually from zero at the critical point (rather than jumping suddenly). This benign, progressive onset is favorable for applications because it ensures smooth and reliable actuation of the wrinkle pattern as conditions change.

Active control of surface instabilities via electric fields is a promising strategy in soft materials research. This approach aligns with major efforts in morphing soft robotic components, flexible and stretchable electronics, smart surface engineering, and bioinspired interfaces, where on-demand reconfigurability is a must. Here, we combine theoretical modeling and finite element simulations to elucidate the electro-mechanical instability of a soft dielectric film bonded to a hyperelastic substrate, addressing this timely challenge from both fundamental and computational perspectives. Our findings not only shed light on the mechanics of electrically induced wrinkling but also demonstrate how electric fields can be exploited to actively tune surface patterns, thereby broadening the design space for functional soft materials.

## 2. Materials and methods

### 2.1. Setup

The system consists of a semi-infinite elastic substrate and an elastic dielectric film glued on its surface. In the initial undeformed configuration, the substrate and the film occupy the  $X_2 > 0$  and  $-H < X_2 < 0$  regions, respectively, where  $(X_1, X_2, X_3)$  are the Cartesian coordinates and  $H$  is the film thickness. In the current deformed configuration, the film-substrate system is deformed homogeneously along the principal directions of stretch  $x_i$  (parallel to the  $X_i$ ), with corresponding stretch ratios  $\lambda_i$ . Hence, the current

film thickness is  $h = \lambda_2 H$ . The film and substrate are perfectly bonded and both are incompressible so that they undergo the same deformation and  $\lambda_1 \lambda_2 \lambda_3 = 1$ .

The deformation is due to the application of an electric field inside the film, generated by the potential difference  $V$  between two flexible electrodes coated on its top and bottom faces. In general, a soft dielectric film expands in its plane under a voltage. Here, for simplicity and to make connections with known results, we focus on the plane-strain deformation  $\lambda_1 = \lambda$ ,  $\lambda_2 = \lambda^{-1}$ ,  $\lambda_3 = 1$ , which is achieved by confining the system between two fixed, lubricated, rigid plates, normal to the  $x_3$ -direction, and applying external forces in the  $x_1$ -direction (see Fig. 2b; note that in Appendix A, we treat the general triaxial case).

To characterize the materials, we choose the neo-Hookean model for the hyperelastic substrate and the neo-Hookean ideal dielectric model for the film, so that their *total* free energy density functions take the form

$$W_s = \frac{1}{2} \mu_s (\lambda^2 + \lambda^{-2} - 2), \quad W_f = \frac{1}{2} \mu_f (\lambda^2 + \lambda^{-2} - 2) - \frac{1}{2} \varepsilon \lambda^2 (V/H)^2, \quad (1)$$

respectively, where the  $\mu_i$  ( $i = s, f$ ) are the initial shear moduli in the undeformed configuration, and  $\varepsilon$  is the film's electric permittivity, which remains unaffected by the deformation. The subscripts  $s$  and  $f$  refer to the physical quantities of the substrate and film, respectively.

When the film is under voltage  $V$ , the whole film-substrate system deforms homogeneously. Because the top surface at  $x_2 = -h$  is free of electro-elastic traction, the normal component of the total stress vanishes there. By continuity, it also vanishes at the perfectly bonded interface. Then, the following *total* Cauchy stress component along the  $x_1$ -direction is required to keep the plane-strain deformation in the film under voltage  $V$  and stretch  $\lambda$  (Su et al., 2018):  $\sigma_f = \lambda W'_f(\lambda) = \mu_f (\lambda^2 - \lambda^{-2}) - \varepsilon \lambda^2 (V/H)^2$ . It follows that at equilibrium, when no lateral traction is applied along  $x_1$  (i.e.,  $\sigma_f = 0$ ), the corresponding stretch  $\lambda_0$  and voltage  $V_0$  are linked as (Su et al., 2019a)

$$\lambda_0 = (1 - \bar{E}_0^2)^{-1/4}, \quad \text{or} \quad \bar{E}_0 = \sqrt{1 - \lambda_0^{-4}}, \quad (2)$$

where  $\bar{E}_0 = \sqrt{\varepsilon/\mu_f} (V_0/H)$  is a non-dimensional measure of the voltage at this equilibrium.

## 2.2. Exact bifurcation

To model the small-amplitude wrinkles appearing at the onset of linearized instability, we assume sinusoidal variations in the  $x_1$ -direction with wavelength  $2\pi/k$ , where  $k$  is the wrinkling wavenumber, and introduce the generalized, non-dimensional displacement-traction vector  $\eta(kx_2) = [U_1, U_2, \Delta, S_{21}, S_{22}, \Phi]^T$ , where  $U_1, U_2$  are the components of the incremental mechanical displacement vector,  $\Delta$  is a measure of the incremental electric displacement in the  $x_2$ -direction,  $S_{21}$  and  $S_{22}$  are the components of a measure of the incremental mechanical traction, and  $\Phi$  is a measure of the incremental electric potential (all quantities depend on the variable  $kx_2$  only).

In the film and substrate, the incremental equations of equilibrium can be formulated as a first-order differential equation,  $\eta' = iN\eta$ , where  $i = \sqrt{-1}$  is the imaginary unit, the prime denotes differentiation with respect to  $kx_2$ , and  $N$  is the (constant) Stroh matrix, with components given explicitly in Appendix A. It is then straightforward to solve the boundary value problem (decay condition as  $x_2 \rightarrow \infty$ , continuity of  $\eta$  at the interface  $x_2 = 0$ , and the conditions of zero traction and a constant applied voltage on the top surface  $x_2 = -h$ ). As shown in Appendix A, the exact bifurcation equation can be put into the compact form

$$\det (\mathbf{Z}_f - r\mathbf{Z}_s) = 0, \quad (3)$$

where  $r = \mu_s/\mu_f$  is the substrate-to-film *stiffness ratio*, and  $\mathbf{Z}_f$  and  $\mathbf{Z}_s$  denote the impedance matrices of the film and substrate, respectively. This equation depends only on the non-dimensional quantities  $r$ ,  $\lambda$ ,  $kh$ , and  $\bar{E}_L = \sqrt{\varepsilon/\mu_f} (V/H)$ .

We may then plot the dispersion curves (also referred to as bifurcation curves) for a given stiffness ratio: either the  $\lambda - kh$  curves when we are interested in wrinkling instability under an increasing mechanical compression for a given voltage, or the  $\bar{E}_L - kh$  curves when we focus on an increasing electric load for a given stretch. Typically, these curves exhibit an extremum: a maximum  $\lambda = \lambda_{cr}$  in the former case, a minimum  $\bar{E}_L = \bar{E}_L^{cr}$  in the latter case (although not always), see Section 3. These extrema are the sought critical stretches and critical voltages of primary bifurcation.

## 2.3. Approximate bifurcation and critical fields

In the Results section, we show that the critical fields occur in the early part of the  $kh$  span, typically when  $kh < 2$ . Moreover, the critical value  $(kh)_{cr}$  decreases as the dielectric film becomes stiffer than the substrate (i.e.,  $r$  is small). It thus makes sense to seek Taylor series expansions of the bifurcation condition (3). As detailed in Appendix A, we expand the relationship  $\eta(-kh) = \exp(-ikhN)\eta(0)$  up to the sixth power in  $kh$ , apply the incremental boundary conditions, and observe that the resultant approximate expansion captures accurately the extrema corresponding to the critical wrinkling values in the early parts of the bifurcation curves.

Furthermore, under the assumption that the film is significantly stiffer than the substrate (i.e.,  $r$  is small, of order  $(kh)^3$ ), we perform an asymptotic analysis based on the sixth-order approximate expansion to obtain explicit asymptotic expressions in  $kh$  for the stretch  $\lambda$  and the voltage  $\bar{E}_L$ . Then we can derive asymptotic expansions of the critical wavenumber  $(kh)_{cr}$  and critical loads  $\lambda_{cr}$  (stretch) and  $\bar{E}_L^{cr}$  (voltage) explicitly by finding the first extremum of the  $kh$ -polynomial asymptotic expressions for the stretch  $\lambda$  and voltage  $\bar{E}_L$ . Each of these critical quantities can be expressed in an  $r^{1/3}$  power series, allowing us to extend the scaling laws of Allen (1969) to electro-elasticity. The detailed derivation is presented in Appendix B, with explicit asymptotic expansions provided in Section 3.3.

## 2.4. Numerical post-buckling analysis

Complex nonlinear behaviors may be expected beyond the bifurcation, such as secondary bifurcations, period doubling, and self-contact folding (Brau et al., 2011; Fu and Cai, 2015). Here, we rely on the finite element method to investigate the post-buckling behavior of the soft dielectric film-substrate system.

We use a quasi-incompressible formulation of the problem to avoid element locking, with the following energy functionals for the substrate and the dielectric film, respectively,

$$\mathcal{E}_s[\mathbf{u}] = \int_{\mathcal{B}_s} \widehat{W}_s(\mathbf{F}) dV, \quad \mathcal{E}_f[\mathbf{u}, \varphi] = \int_{\mathcal{B}_f} \widehat{W}_f(\mathbf{F}, \mathbf{E}_L) dV, \quad (4)$$

where  $\mathbf{u}$  and  $\varphi$  are the mechanical displacement vector and the electric-potential field,  $\mathbf{F}$  is the deformation gradient (two-point) tensor,  $\mathbf{E}_L = -\text{Grad}\varphi$  is the Lagrangian electric field vector,  $\widehat{W}_s$  and  $\widehat{W}_f$  are the strain energy densities of the substrate and dielectric film, respectively, and  $\mathcal{B}_s$  and  $\mathcal{B}_f$  are the domains of the substrate and film.

For the nearly incompressible versions of the total energy densities (1), we take

$$\widehat{W}_s = \frac{1}{2}\mu_s(I_1^* - 3) + \frac{1}{2}K_s(\ln J)^2, \quad \widehat{W}_f = \frac{1}{2}\mu_f(I_1^* - 3) + \frac{1}{2}K_f(\ln J)^2 - \frac{1}{2}\epsilon J \mathbf{E} \cdot \mathbf{E}, \quad (5)$$

where  $\mathbf{E} = \mathbf{F}^{-T}\mathbf{E}_L$  is the Eulerian electric field vector,  $J = \det(\mathbf{F})$ ,  $I_1^* = J^{-2/3}\text{tr}(\mathbf{F}^T\mathbf{F})$  and the  $K_i$  ( $i = s, f$ ) are the initial bulk moduli (chosen to be much larger than the  $\mu_i$ ). It can be shown that the stationary points of the total energy functional  $\mathcal{E} = \mathcal{E}_f + \mathcal{E}_s$  correspond to equilibrium configurations of the system, see Toupin (1956) and Dorfmann and Ogden (2014). To approximate the fully incompressible case, we set  $K_i = 500\mu_i$  ( $i = s, f$ ), so that the initial Poisson ratio is

$$\nu_i = \frac{3K_i - 2\mu_i}{2(3K_i + \mu_i)} \simeq 0.499, \quad (i = s, f). \quad (6)$$

The system is modeled by using a rectangular computational domain  $[0, L] \times [D, -H]$ , where the depth  $D$  of the substrate is large compared to the film thickness  $H$  (specifically, we set  $D = 30H$ ), and the length  $L$  is chosen as half of the wavelength of the wrinkling pattern. On the left and right sides of the domain, we impose symmetry boundary conditions to mimic the infinite layered half-space. We use a triangular structured mesh in the dielectric film (with at least ten elements along the width), while the mesh is unstructured in the substrate, with a coarser mesh as we move away from the film-substrate interface.

To track the bifurcated branch, an arclength continuation algorithm is used, as described in Su et al. (2023), where either the stretch  $\lambda$  or the non-dimensional voltage  $\bar{E}_L$  are used as the control parameter. However, the handling of a control parameter that enters the boundary conditions is not straightforward using the arclength continuation algorithm. To avoid this issue, we split the displacement field additively as  $\mathbf{u} = \mathbf{u}_h + \mathbf{u}_i$ , where  $\mathbf{u}_h = (\lambda - 1)X_1\hat{\mathbf{l}}_1 + (\lambda^{-1} - 1)X_2\hat{\mathbf{l}}_2$  is the displacement field corresponding to the homogeneous deformation, and  $\mathbf{u}_i$  is the inhomogeneous displacement field corresponding to the wrinkles, with  $\hat{\mathbf{l}}_1$  and  $\hat{\mathbf{l}}_2$  representing the unit basis vectors of the initial undeformed configuration. As  $\mathbf{u}_h$  is known, we can solve the problem with respect to  $\mathbf{u}_i$  to reconstruct the full displacement field. A similar splitting is performed for the electric potential, i.e.,  $\varphi = \varphi_h + \varphi_i$  with  $\varphi_h = -VX_2$ . The Dirichlet boundary conditions on  $\mathbf{u}_i$  and  $\varphi_i$  are homogeneous, facilitating a more straightforward implementation of the arclength method (see also Riccobelli et al. (2024)). A piecewise quadratic polynomial basis is employed for the inhomogeneous displacement, while a piecewise linear polynomial basis is used for the electric potential. A small imperfection is applied to the top surface to trigger the instability. The amplitude of the imperfection is the smallest value for which the bifurcation is detected by the arclength method (i.e.,  $5 \times 10^{-5}H$ , unless explicitly stated otherwise).

We solve the problem using the finite element method implemented in FEniCS (Alnæs et al., 2015), which allows for automatic differentiation of the weak form and efficient assembly of the linear system. We obtain the solution by solving a Newton-Raphson problem, where the Jacobian matrix is computed using the automatic differentiation library UFL. For the continuation algorithm, we use the arclength method implemented in BiFEniCS.

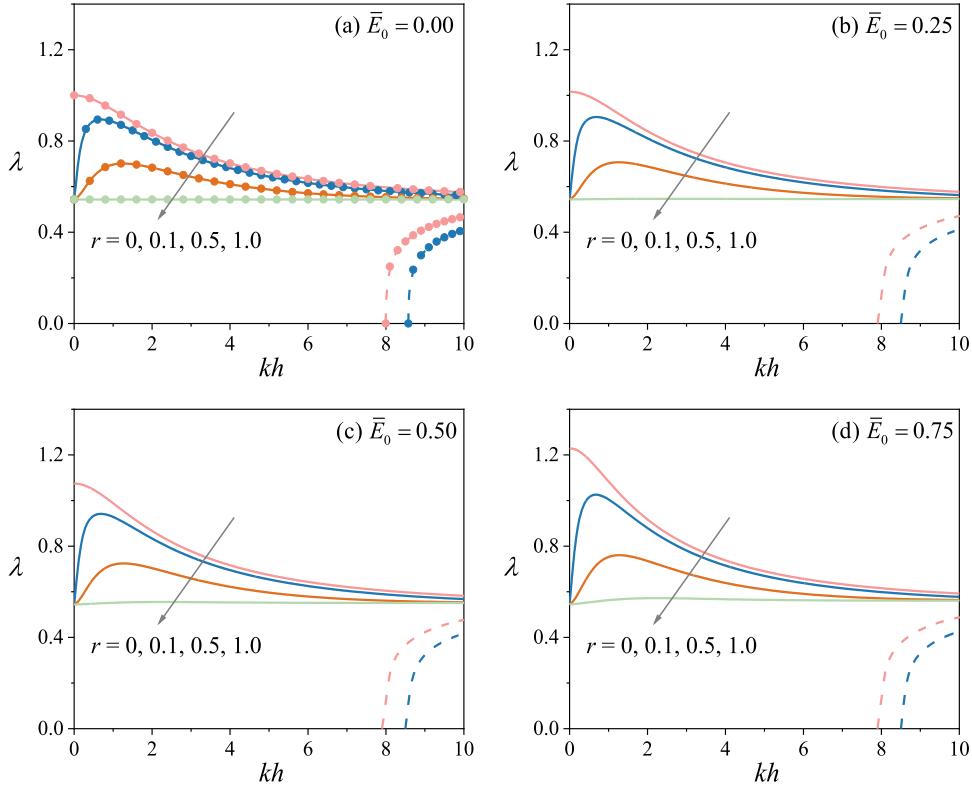
## 3. Results and discussion

### 3.1. Critical bifurcation stretch for a prescribed electric field

First we consider the scenario where the applied voltage is fixed at its initial traction-free equilibrium value  $V_0$ , and the film-substrate system is deformed homogeneously under the action of a uniaxial stress along  $x_1$ . Hence, the total Cauchy stresses  $\sigma_f = \mu_f(\lambda^2 - \lambda^{-2}) - \epsilon\lambda^2(V_0/H)^2$  and  $\sigma_s = \mu_s(\lambda^2 - \lambda^{-2})$  are applied to the dielectric film and the substrate, respectively.

Fig. 3 shows the bifurcation curves of stretch  $\lambda$  against wavenumber  $kh$  for different shear modulus ratios  $r = \mu_s/\mu_f$  under four given non-dimensional voltages  $\bar{E}_0 = 0, 0.25, 0.5, 0.75$ . When  $\bar{E}_0 = 0$ , we have excellent agreement with Cai and Fu (1999), who used the classical displacement solution method for a coated hyperelastic half-space, confirming the effectiveness of the surface impedance method, see Fig. 3(a). All bifurcation curves, except for the  $r = 0$  one, start at  $\lambda = 0.5437$ , the Biot critical stretch of instability for an elastic neo-Hookean half-space, which corresponds to  $kh = 0$  (vanishing film thickness). When  $r = 0$ , there is no substrate; then, in the  $kh \rightarrow 0$  limit, the dielectric film is infinitesimally thin and wrinkles immediately once  $\sigma_f$  is applied. Hence, the limit is found by solving  $\sigma_f = 0$ , which according to Eq. (2), gives the limits  $\lambda = 1.000, 1.016, 1.075, 1.230$  for  $\bar{E}_0 = 0, 0.25, 0.5, 0.75$ , respectively. Conversely, in the  $kh \rightarrow \infty$  limit, the soft dielectric film becomes a semi-infinite ideal dielectric, and all bifurcation curves tend to the root of

$$\lambda^6 + \lambda^4 + 3\lambda^2 - 1 = \lambda^4(1 + \lambda^2)\bar{E}_0^2, \quad (7)$$



**Fig. 3.** Bifurcation curves of stretch  $\lambda$  as a function of  $kh$  with different substrate-to-film shear modulus ratios  $r = \mu_s/\mu_f = 0, 0.1, 0.5, 1.0$  under four non-dimensional voltages applied to the soft dielectric film, subjected to a plane-strain load: (a)  $\bar{E}_0 = 0$ ; (b)  $\bar{E}_0 = 0.25$ ; (c)  $\bar{E}_0 = 0.5$ ; (d)  $\bar{E}_0 = 0.75$ . Solid curves: antisymmetric-dominated modes; Dashed curves: symmetric-dominated modes. The onset of instability occurs at the maximum of the bifurcation curve, always corresponding to an antisymmetric-dominated mode. In Fig. 3(a), solid lines show the proposed surface impedance results, and symbols indicate the displacement solution predictions (Cai and Fu, 1999).

which corresponds to the surface instability criterion in plane strain (see Appendix A for more general formulas and details). It yields  $\lambda \rightarrow 0.5437, 0.5454, 0.5508, 0.5607$  when  $\bar{E}_0 = 0, 0.25, 0.5, 0.75$ , respectively.

Between these two limits, the bifurcation curve goes through a maximum, which determines the critical stretch  $\lambda_{\text{cr}}$  and critical wavenumber  $(kh)_{\text{cr}}$ . We collected these critical values for six given voltages and two stiffness ratios in Table 1. When there is no applied voltage ( $\bar{E}_0 = 0$ ), the values of the critical compressive strain are  $\epsilon_w = 1 - \lambda_{\text{cr}} = 0.165$  and 0.052 for  $r = 1/5$  and  $1/30$ , respectively, in reasonable agreement with Cao and Hutchinson (2012). We observe from Fig. 3 and Table 1 that, for a fixed modulus ratio  $r$ , the critical stretch  $\lambda_{\text{cr}}$  monotonically increases with increasing applied voltage  $\bar{E}_0$ , and that, for a prescribed voltage,  $\lambda_{\text{cr}}$  also increases as  $r$  decreases. While the applied voltage exerts only a negligible influence on the critical wavenumber  $(kh)_{\text{cr}}$ , the latter progressively decreases with decreasing modulus ratio.

To further elucidate the influence of the applied voltage  $\bar{E}_0$  on  $\lambda_{\text{cr}}$  and  $(kh)_{\text{cr}}$ , we seek their asymptotic expansions in terms of  $r$  and  $\bar{E}_0$ . But prior to that, Fig. 4 shows that the bifurcation curves of stretch  $\lambda$  versus  $kh$  predicted by the sixth-order Taylor approximate solution (A.8) agrees remarkably well with the exact bifurcation equation (3) over a broad range of  $kh$ , thereby validating its accuracy in capturing the extrema of the bifurcation curves and providing a solid foundation for an asymptotic analysis of the critical parameters  $\lambda_{\text{cr}}$  and  $(kh)_{\text{cr}}$ .

Then we use the sixth-order approximation to perform the asymptotic analysis (see Appendix B and Section 3.3) and derive the explicit leading-order correction for the relative extension of wrinkling instability due to electro-mechanical loading, as

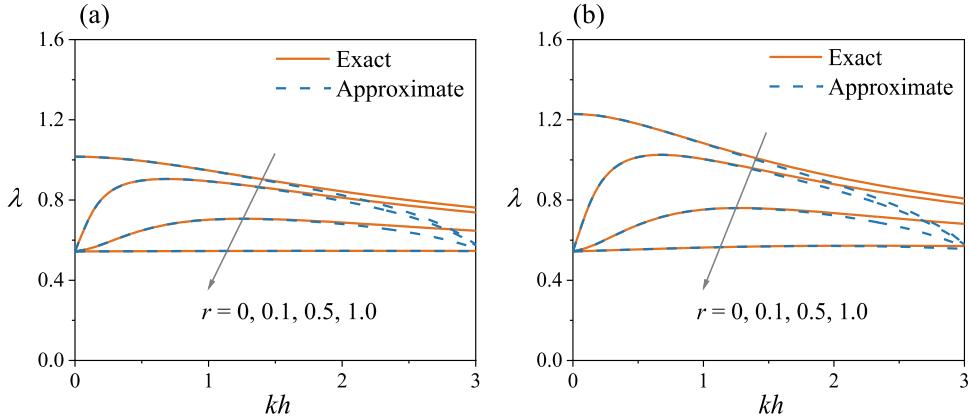
$$\epsilon_w = \left| \frac{\lambda_{\text{cr}} - \lambda_0}{\lambda_0} \right| = \frac{1}{4} \frac{1}{1 - \bar{E}_0^2} \left( \frac{1 + \sqrt{1 - \bar{E}_0^2}}{2} 3 \frac{\mu_s}{\mu_f} \right)^{2/3}, \quad (8)$$

where we recall that  $\bar{E}_0 \leq 1$ . This expression recovers the classical  $2/3$  power scaling law with respect to the modulus ratio  $r = \mu_s/\mu_f$  for the purely elastic case, as originally reported by Allen (1969), while additionally revealing that the multiplicative factor associated with  $\bar{E}_0$  is a monotonically increasing function of the applied voltage. This indicates that a higher initial voltage permits a larger relative compressive strain, quantified as  $\lambda_0 - \lambda_{\text{cr}}$ , to trigger instability. In other words, once the initial traction-free voltage  $\bar{E}_0$  is applied, the system can sustain greater homogeneous deformation before the onset of instability, thereby postponing the emergence

**Table 1**

Critical stretch and wavenumber values, together with the classification of bifurcation type (supercritical or subcritical), for different electric loadings  $\bar{E}_0$  and shear modulus ratios  $r$ . The notation “—” indicates cases where the asymptotic solution is inapplicable. Supercripts “num” and “asymp” represent the critical values calculated by finite element numerical simulations and the asymptotic expansion expressions (14) and (15), respectively.

	Value for $\bar{E}_0 = 0$	Value for $\bar{E}_0 = 0.3$	Value for $\bar{E}_0 = 0.6$	Value for $\bar{E}_0 = 0.75$	Value for $\bar{E}_0 = 0.9$	Value for $\bar{E}_0 = 1.0$
<b>Critical values when <math>r = 1/5</math></b>						
$\lambda_{\text{cr}}$	0.8354	0.8481	0.8933	0.9372	1.0119	1.1005
$\lambda_{\text{cr}}^{\text{num}}$	0.8353	0.8481	0.8936	0.9378	1.0134	1.1085
$\lambda_{\text{cr}}^{\text{asymp}}$	0.8569	0.8756	0.9605	1.1097	2.3088	—
$(kh)_{\text{cr}}$	0.88	0.88	0.88	0.88	0.86	0.86
$(kh)_{\text{cr}}^{\text{asymp}}$	0.87	0.88	0.89	0.91	0.99	—
super/subcritical	super-	super-	super-	super-	super-	sub-
<b>Critical values when <math>r = 1/30</math></b>						
$\lambda_{\text{cr}}$	0.9482	0.9674	1.0401	1.1190	1.2822	1.5874
$\lambda_{\text{cr}}^{\text{num}}$	0.9486	0.9680	1.0408	1.1200	1.2842	1.6013
$\lambda_{\text{cr}}^{\text{asymp}}$	0.9493	0.9689	1.0435	1.1277	1.3548	—
$(kh)_{\text{cr}}$	0.46	0.46	0.46	0.46	0.44	0.44
$(kh)_{\text{cr}}^{\text{asymp}}$	0.47	0.47	0.46	0.46	0.45	—
super/subcritical	super-	super-	super-	super-	super-	sub-



**Fig. 4.** Exact and approximate (sixth-order) bifurcation curves of stretch  $\lambda$  as functions of  $kh$  for different shear modulus ratios  $r = \mu_s/\mu_f$  under two non-dimensional voltages, (a)  $\bar{E}_0 = 0.25$  and (b)  $\bar{E}_0 = 0.75$ , showing that the approximations capture the critical points accurately, thus enabling asymptotic expansions of  $\lambda_{\text{cr}}$  and  $(kh)_{\text{cr}}$  in terms of  $r$  and  $\bar{E}_0$ .

of wrinkles. Note that using Eq. (2), the result can equivalently be expressed in terms of the initial traction-free pre-stretch  $\lambda_0$  as

$$\epsilon_w = \frac{\lambda_0^4}{4} \left( \frac{1 + \lambda_0^{-2}}{2} 3 \frac{\mu_s}{\mu_f} \right)^{2/3}. \quad (9)$$

Similarly, the scaling relation governing the leading-order correction to the critical wavenumber is derived as

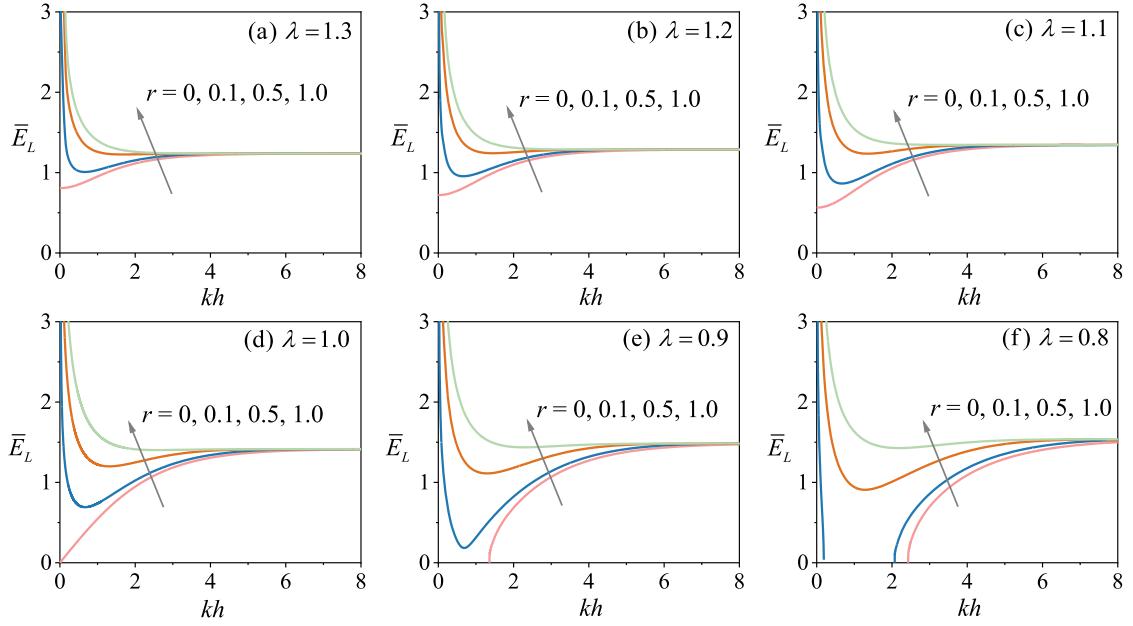
$$(kh)_{\text{cr}} = \left( \frac{1 + \sqrt{1 - \bar{E}_0^2}}{2} 3 \frac{\mu_s}{\mu_f} \right)^{1/3} = \left( \frac{1 + \lambda_0^{-2}}{2} 3 \frac{\mu_s}{\mu_f} \right)^{1/3}, \quad (10)$$

see Appendix B and Section 3.3, which provides details and further approximations and expansions.

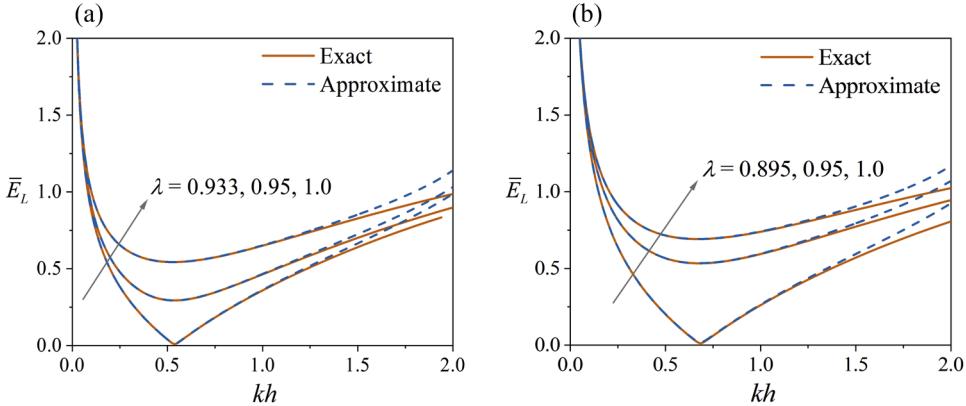
### 3.2. Critical bifurcation electric voltage for a fixed pre-stretch

Conversely, we may hold the pre-stretch at an initial fixed value  $\lambda$  and observe wrinkling as the applied voltage  $V$  changes. In that scenario, the pre-load is achieved by applying the uniaxial Cauchy stresses  $\sigma_f = \mu_f(\lambda^2 - \lambda^{-2}) - \varepsilon \lambda^2 (V/H)^2$  in the dielectric film and  $\sigma_s = \mu_s(\lambda^2 - \lambda^{-2})$  in the substrate.

The bifurcation curves of the non-dimensional voltage  $\bar{E}_L = \sqrt{\varepsilon/\mu_f} (V/H)$  as functions of the wavenumber  $kh$  are presented in Fig. 5 for various shear modulus ratios  $r = \mu_s/\mu_f$  under six prescribed pre-stretch values  $\lambda = 1.3, 1.2, 1.1, 1.0, 0.9, 0.8$ . The results reveal



**Fig. 5.** Bifurcation curves of the non-dimensional voltage  $\bar{E}_L$  as a function of  $kh$  for different stiffness ratios  $r = 0, 0.1, 0.5, 1.0$  and six fixed pre-stretches: (a–c) extensile stretches  $\lambda = 1.3, 1.2, 1.1$ ; (d–f) contractile stretches  $\lambda = 1.0, 0.9, 0.8$ . The minima correspond to the critical voltage  $\bar{E}_L^{cr}$  and critical wavenumber  $(kh)^{cr}$ . For a sufficiently contractile stretch ( $\lambda < \lambda_c^0$ ) and a sufficiently stiff film ( $r < r_c^0$ ), the system wrinkles before the application of voltage, as shown in Fig. 5(f) for  $r = 0.1$ , for example.

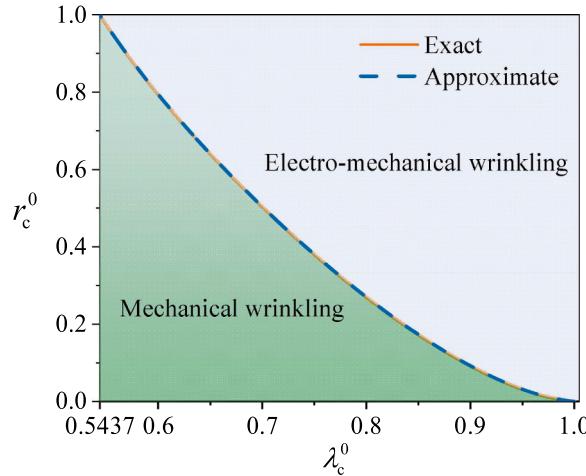


**Fig. 6.** Exact and approximate (sixth-order) bifurcation curves of voltage  $\bar{E}_L$  as functions of  $kh$  for different pre-stretches  $\lambda$  and two shear modulus ratios, (a)  $r = 0.05$  and (b)  $r = 0.1$ , demonstrating that the approximations accurately capture the critical points, thus enabling asymptotic expansions of  $\bar{E}_L^{cr}$  and  $(kh)^{cr}$  in terms of  $r$  and  $\lambda$ . For a pre-stretch  $\lambda$  marginally exceeding  $\lambda_c^0 = 0.933$  (a) and  $0.895$  (b), even a small applied voltage is sufficient to induce instability.

that the bifurcation curves of  $\bar{E}_L$  with respect to  $kh$  generally exhibit non-monotonic behavior, except in the special case of  $r = 0$ , where the curve increases monotonically. However, all bifurcation curves exhibit a minimum, corresponding to the critical voltage  $\bar{E}_L^{cr}$  of primary interest. Moreover, Fig. 5 reveals that both decreasing pre-stretch  $\lambda$  and reducing modulus ratio  $r$  lead to a progressive decline in the critical voltage  $\bar{E}_L^{cr}$ , thereby indicating an increased susceptibility of the system to wrinkling instability. In particular, if the pre-stretch is sufficiently contractile ( $\lambda < 1$ ) and the film is sufficiently stiff ( $r$  small), we expect that wrinkling may occur for small values of the voltage loading, which is confirmed by the trend in Fig. 5(e) and (f).

To investigate the effect of the applied pre-stretch  $\lambda$  on the critical voltage  $\bar{E}_L^{cr}$  and the critical wavenumber  $(kh)^{cr}$ , we seek their asymptotic representations with respect to  $r$  and  $\lambda$ . Again, the sixth-order Taylor expansion leads to an excellent agreement with the exact bifurcation condition across a wide interval of  $kh$ , establishing a rigorous basis for the asymptotic characterization of the critical quantities  $\bar{E}_L^{cr}$  and  $(kh)^{cr}$ , see Fig. 6.

Moreover, Fig. 6 reveals that, for a prescribed modulus ratio  $r$ , a decrease in the applied pre-stretch  $\lambda$  leads to a gradual reduction in the corresponding critical voltage  $\bar{E}_L^{cr}$ , a trend consistent with the behavior previously identified in Fig. 5. Particularly, the critical



**Fig. 7.**  $\lambda_c^0 - r_c^0$  critical curve corresponding to a vanishing critical voltage (i.e., the purely elastic limit), below which the film-substrate system wrinkles mechanically prior to the application of a voltage, whereas above which a finite applied voltage is required to trigger wrinkling instability.

voltage  $\bar{E}_L^{\text{cr}}$  may eventually reach zero for  $r = 0.05$ ,  $\lambda \simeq 0.933$ , and  $r = 0.1$ ,  $\lambda \simeq 0.895$ , at which point we recover the critical values for the stretch and wavenumber of a purely elastic film-substrate system (Cai and Fu, 1999; Cao and Hutchinson, 2012). When  $r$  or  $\lambda$  is further reduced, the incremental analysis breaks down, and the negative value of the minimum has no physical meaning (e.g., the case  $\lambda = 0.8$  and  $r = 0.1$  in Fig. 5(f)).

Fig. 7 shows the domain in the  $\lambda - r$  plane where the soft dielectric film-substrate system can be expected to wrinkle under an applied voltage. The demarcation curve, referred to as the  $\lambda_c^0 - r_c^0$  critical curve, corresponds to a vanishing critical electric field (i.e., the purely elastic limit). This critical curve is found by solving the exact or the sixth-order approximate bifurcation condition when  $\bar{E}_L^{\text{cr}} = 0$ , and behaves asymptotically as

$$\lambda_c^0 = 1 - \frac{1}{4}(3r_c^0)^{2/3}, \quad (11)$$

according to Eq. (8) written at  $\bar{E}_0 = 0$  and  $\lambda_0 = 1$ , in agreement with Cai and Fu (1999). In Fig. 7, we denote by  $(\lambda_c^0, r_c^0)$  the coordinates of points on that critical curve, with  $(kh)_c^0$  representing the corresponding critical wavenumber. Parameter combinations of pre-stretch and stiffness ratio underneath that critical curve lead to wrinkling prior to the application of any voltage, whereas those above the critical curve require a finite applied voltage to trigger wrinkling instability.

We also employ the sixth-order approximation and perform the asymptotic analysis (see Appendix B and Section 3.3) to obtain an explicit expression for the leading-order correction to the squared critical voltage in terms of  $r$  and  $\lambda$  as,

$$(\bar{E}_L^{\text{cr}})^2 = 1 - \lambda^{-4} + \left( \frac{1 + \lambda^{-2}}{2} 3r \right)^{2/3}, \quad (12)$$

provided the values of  $(\lambda, r)$  are not in the shaded area of Fig. 7. This is equivalent to  $(\bar{E}_L^{\text{cr}})^2 \geq 0$ , or, by expansion for small  $r$ ,  $\lambda \geq 1 - (1/4)(3r)^{2/3}$ , in agreement with Eq. (11).

We collected the critical values of voltage and wavenumber for six given pre-stretches and two stiffness ratios in Table 2. It shows that a reduction in the pre-stretch  $\lambda$  or a decrease in the modulus ratio  $r$  diminishes the critical voltage  $\bar{E}_L^{\text{cr}}$ , thereby indicating an enhanced propensity of the soft dielectric film-substrate system to undergo wrinkling instability, as mentioned earlier. In addition, under pre-compression ( $\lambda < 1$ ), only a tiny voltage is required to trigger wrinkling when the dielectric film is much stiffer than the substrate. We also see that an applied (albeit larger) voltage can render the system unstable when it is pre-elongated ( $\lambda > 1$ ), because the film tends to expand in its plane under the applied voltage, which is prevented when  $\lambda$  is fixed, eventually leading to wrinkles.

### 3.3. Asymptotic expansions for high-contrast stiffness ratios

The numerical root-finding procedure for the exact bifurcation equation (3) incurs a heavy computational cost, such that asymptotic expansions may provide a much-needed rapid alternative way to find the critical values of stretch and voltage.

For small  $r$  and  $kh$ , and under the assumption that  $r$  is of order  $(kh)^3$ , we show in Appendix B how a fourth-order series expansion can be derived for the stretch through asymptotic analysis,

$$\begin{aligned} \frac{\lambda}{\lambda_0} = 1 - \frac{1}{4}\lambda_0^2(1 + \lambda_0^2)\left(\frac{r}{kh}\right) - \frac{1}{12}\lambda_0^4(kh)^2 + \frac{1}{4}(\lambda_0^2 - 1)r \\ + \frac{1}{48}(\lambda_0^2 - 1)(3 + 7\lambda_0^2 + 8\lambda_0^4 + 5\lambda_0^6)r(kh) + \frac{1}{1440}\lambda_0^4(2 + 37\lambda_0^4)(kh)^4 \\ + \frac{1}{32}(2 - 4\lambda_0^2 + 3\lambda_0^4 + 6\lambda_0^6 + 5\lambda_0^8)\left(\frac{r}{kh}\right)^2, \end{aligned} \quad (13)$$

**Table 2**

Critical voltage and wavenumber values, together with the classification of bifurcation type (supercritical or subcritical), for different mechanical loadings  $\lambda$  and shear modulus ratios  $r$ . The notation “—” indicates cases where the wrinkling has occurred prior to the application of the voltage. Subscripts “num” and “asym” represent the critical values calculated by finite element numerical simulations and the asymptotic expansion expressions (19) and (20), respectively.

	Value for $\lambda = 0.85$	Value for $\lambda = 0.9$	Value for $\lambda = 0.95$	Value for $\lambda = 1.0$	Value for $\lambda = 1.1$	Value for $\lambda = 1.2$
<b>Critical values when <math>r = 1/5</math></b>						
$\bar{E}_L^{\text{cr}}$	0.3207	0.6280	0.7825	0.8814	0.9996	1.0535
$\bar{E}_{\text{num}}^{\text{cr}}$	0.3214	0.6278	0.7819	0.8808	0.9913	1.0038
$\bar{E}_{\text{asym}}^{\text{cr}}$	0.3962	0.6605	0.8034	0.8967	1.0098	1.0724
$(kh)^{\text{cr}}$	0.88	0.87	0.87	0.86	0.86	0.86
$(kh)_{\text{asym}}^{\text{cr}}$	0.82	0.82	0.82	0.82	0.82	0.83
super/subcritical	super-	super-	super-	super-	sub-	sub-
<b>Critical values when <math>r = 1/30</math></b>						
$\bar{E}_L^{\text{cr}}$	—	—	0.0937	0.4729	0.7213	0.8407
$\bar{E}_{\text{num}}^{\text{cr}}$	—	—	0.0949	0.4723	0.7210	0.8407
$\bar{E}_{\text{asym}}^{\text{cr}}$	—	—	0.0985	0.4732	0.7216	0.8409
$(kh)^{\text{cr}}$	—	—	0.47	0.46	0.45	0.45
$(kh)_{\text{asym}}^{\text{cr}}$	—	—	0.47	0.46	0.45	0.44
super/subcritical	—	—	super-	super-	super-	super-

where  $\lambda_0 = (1 - \bar{E}_0^2)^{-1/4}$  and we neglect terms of order  $(kh)^6$  and higher. Then, by differentiating Eq. (13) with respect to  $kh$ , we find where the stretch is (locally) maximized in the bifurcation curve. Disregarding contributions of order  $r^{4/3}$  and higher, the expression for the critical wavenumber is obtained as

$$(kh)_{\text{cr}} = \left( \frac{1 + \lambda_0^{-2}}{2} 3r \right)^{1/3} + \frac{12\lambda_0^{10} + 54\lambda_0^8 + 19\lambda_0^6 + 19\lambda_0^4 - 53\lambda_0^2 - 15}{120\lambda_0^4(1 + \lambda_0^2)} r, \quad (14)$$

and neglecting terms of order  $r^2$  and higher, the critical stretch is given by

$$\frac{\lambda_{\text{cr}}}{\lambda_0} = 1 - \frac{1}{4} \lambda_0^4 \left( \frac{1 + \lambda_0^{-2}}{2} 3r \right)^{2/3} + \frac{1}{4} (\lambda_0^2 - 1) r + \frac{1}{1920} (237\lambda_0^{10} + 354\lambda_0^8 + 139\lambda_0^6 - 176\lambda_0^4 - 98\lambda_0^2 - 60) \left( \frac{2}{\lambda_0(1 + \lambda_0^2)} \right)^{2/3} r (3r)^{1/3}. \quad (15)$$

Given that  $\lambda_0 = (1 - \bar{E}_0^2)^{-1/4} \geq 0$ , the validity of Eqs. (13)–(15) is restricted to the regime  $\bar{E}_0 \leq 1$ . This regime,  $\bar{E}_0 \leq 1$ , also corresponds to the parameter range where we expect wrinkles to arise under a small electric voltage.

In the absence of an applied voltage ( $\bar{E}_0 = 0$ ,  $\lambda_0 = 1$ ), we recover the asymptotic formulas of the hyperelastic film-substrate system (Cai and Fu, 1999; Alawiye et al., 2019) for the stretch,

$$\lambda = 1 - \frac{1}{2} \left( \frac{r}{kh} \right) - \frac{1}{12} (kh)^2 + \frac{3}{8} \left( \frac{r}{kh} \right)^2 + \frac{13}{480} (kh)^4, \quad (16)$$

and for the critical stretch and critical wavenumber,

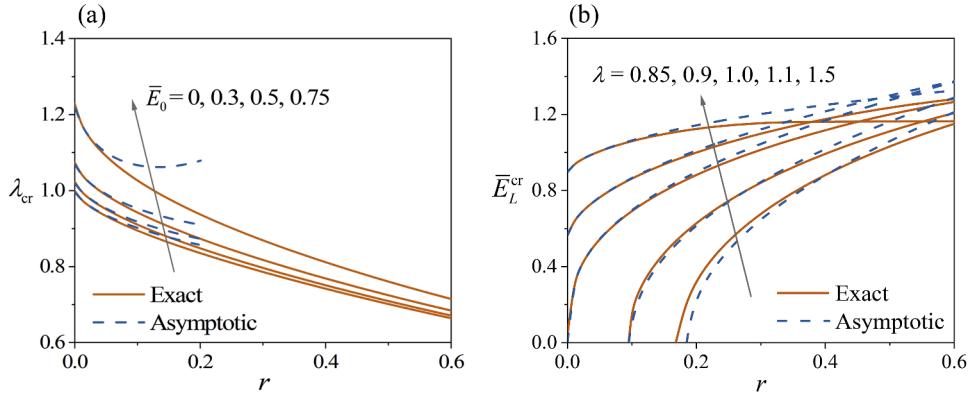
$$\lambda_{\text{cr}} = 1 - \frac{1}{4} (3r)^{2/3} + \frac{33}{160} r (3r)^{1/3}, \quad (kh)_{\text{cr}} = (3r)^{1/3} + \frac{3}{20} r. \quad (17)$$

Similarly, we obtain the asymptotic expansion of the squared non-dimensional voltage in the form

$$\begin{aligned} \bar{E}_L^2 = 1 - \lambda^{-4} + \frac{1}{3} (kh)^2 + (1 + \lambda^{-2}) \left( \frac{r}{kh} \right) + (\lambda^{-4} - \lambda^{-2}) r \\ - \frac{1}{4} (1 - \lambda^{-2})^2 \left( \frac{r}{kh} \right)^2 + \frac{1}{12} (1 + \lambda^{-2})(1 + 3\lambda^{-2}) r (kh) - \frac{1}{180} (1 + 6\lambda^4) (kh)^4. \end{aligned} \quad (18)$$

In the absence of voltage,  $\bar{E}_L = 0$  and we recover Eq. (16). By setting the derivative of Eq. (18) with respect to  $kh$  to zero, we determine the location of the (local) minimum. Neglecting terms of order  $r^2$  and higher, the asymptotic expression for the squared critical voltage is obtained as

$$\begin{aligned} (\bar{E}_L^{\text{cr}})^2 = 1 - \lambda^{-4} + \left( \frac{1 + \lambda^{-2}}{2} 3r \right)^{2/3} - (\lambda^{-2} - \lambda^{-4}) r \\ - \frac{6\lambda^{10} + 12\lambda^8 + 17\lambda^6 - 88\lambda^4 - 49\lambda^2 - 30}{40 \cdot 2^{1/3} \cdot 3^{2/3} \lambda^{14/3} (\lambda^2 + 1)^{2/3}} r^{4/3}, \end{aligned} \quad (19)$$



**Fig. 8.** Variations in critical fields with the shear modulus ratio  $r = \mu_s/\mu_f$ : (a) critical stretch  $\lambda_{\text{cr}}$  for different applied voltages  $\bar{E}_0$ ; (b) Critical voltage  $\bar{E}_L^{\text{cr}}$  for various pre-stretches  $\lambda$ . Solid curves: exact solutions given by Eq. (3); Dashed curves: asymptotic expansions provided by Eqs. (15) and (19). Asymptotic expansions of the critical fields provide a fast alternative to solving the exact bifurcation criterion when the soft dielectric film is much stiffer than the substrate ( $r$  small).

an expression that is valid provided  $\lambda$  is greater than the right-hand side of Eq. (17)<sub>1</sub>. Again, we may verify that when  $\bar{E}_L^{\text{cr}} = 0$ , Eq. (19) is consistent with Eq. (17)<sub>1</sub>. The asymptotic expansion for the corresponding critical wavenumber is

$$(kh)^{\text{cr}} = \left( \frac{1 + \lambda^{-2}}{2} 3r \right)^{1/3} + \frac{12\lambda^{10} + 24\lambda^8 - 11\lambda^6 + 19\lambda^4 - 53\lambda^2 - 15}{120\lambda^4(\lambda^2 + 1)} r. \quad (20)$$

The validity of the asymptotic expansions provided by Eqs. (15) and (19) is illustrated in Fig. 8, exhibiting excellent agreement for small values of  $r$  and reasonable accuracy for moderate  $r$ .

### 3.4. Post-buckling analysis by the finite element method

We now turn to post-buckling analysis. First we plot the non-dimensional amplitude of the free surface wrinkles,  $\Delta y/H$ , as a function of the stretch  $\lambda$  for  $r = 1/5$  and various non-dimensional electric voltages  $\bar{E}_0 = 0.3, 0.6, 0.75$ , see Fig. 9(a). Here,  $\Delta y$  denotes the difference between the maximum and minimum vertical positions of points on the free surface. The resulting diagrams are reminiscent of supercritical pitchfork bifurcations.

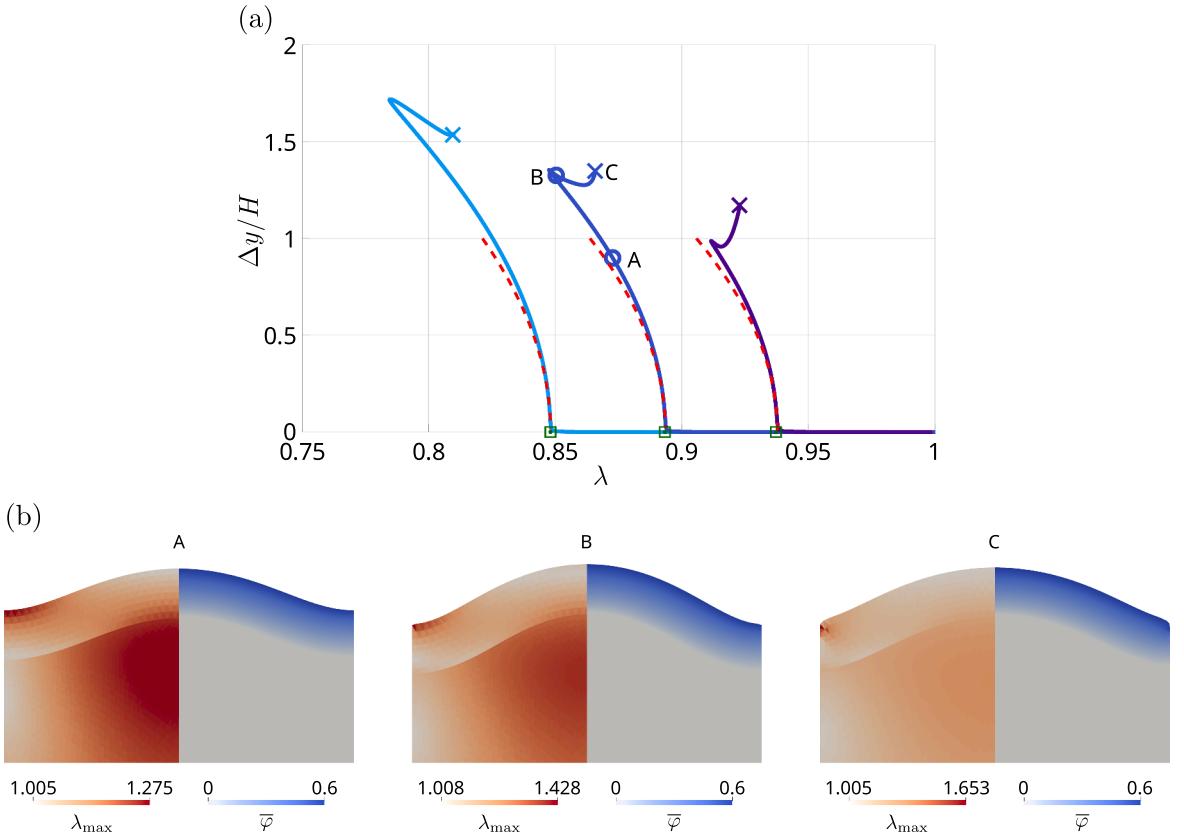
To verify this hypothesis, we fit the following function

$$\frac{\Delta y}{H} = \hat{A} \sqrt{|\lambda_{\text{cr}}^{\text{num}} - \lambda|} \quad (21)$$

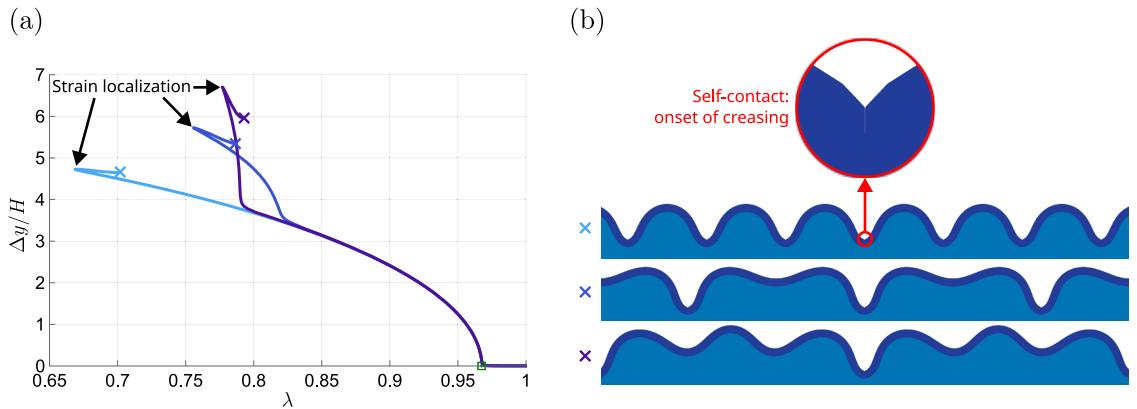
to the finite element numerical data near the bifurcation point. Such a parabolic function represents the amplitude of the wrinkling pattern for a pitchfork bifurcation in the weakly nonlinear regime. From the numerical simulations, we retain only the data satisfying  $0.05H < \Delta y < 0.2H$  in the fitting procedure. This filtering eliminates the influence of strong nonlinear effects and surface imperfections. The upper bound of  $0.2H$  is chosen to ensure optimal fitting across all parameter sets reported in Tables 1 and 2, although in some cases (see Figs. 9 and 11) the range of validity of Eq. (21) extends beyond this limit. Eq. (21) allows to fit the data both in the case of a supercritical and subcritical transition. Indeed, the bifurcation is classified as supercritical if, in the filtered data,  $\lambda < \lambda_{\text{cr}}$ ; otherwise, it is subcritical. In Eq. (21),  $\hat{A}$  modulates the amplitude of the wrinkling close to the bifurcation point, while  $\lambda_{\text{cr}}^{\text{num}}$  represents the numerical threshold of the wrinkling bifurcation. The resulting fit demonstrates excellent agreement with the finite element results near the bifurcation point, see Fig. 9(b). Moreover, the numerically determined critical stretches  $\lambda_{\text{cr}}^{\text{num}}$  closely match the thresholds  $\lambda_{\text{cr}}$  predicted by the linearized stability analysis, with a precision of the order of  $10^{-4}$ , see Table 1. The only exception occurs for the case where  $r = 1/30$  and  $\bar{E}_0 = 1$ , where the discrepancy between the theoretical and numerical thresholds is of the order of  $10^{-2}$ . In this case, the bifurcation is subcritical and abrupt, becoming nonlinear very close to the bifurcation threshold and influencing the fitting procedure.

We note a turning point in all bifurcation diagrams in the fully nonlinear regime. Corresponding to these turning points, the finite element simulations reveal a progressive localization of the deformation, indicating that the strain becomes concentrated in narrow regions near the furrows of the wrinkling pattern rather than remaining uniformly distributed across the surface. This process eventually leads to self-contact (see the deformed configurations corresponding to points A, B, C indicated in the bifurcation diagram in Fig. 9), a transition reminiscent of the creasing onset observed by Hohlfeld and Mahadevan (2011).

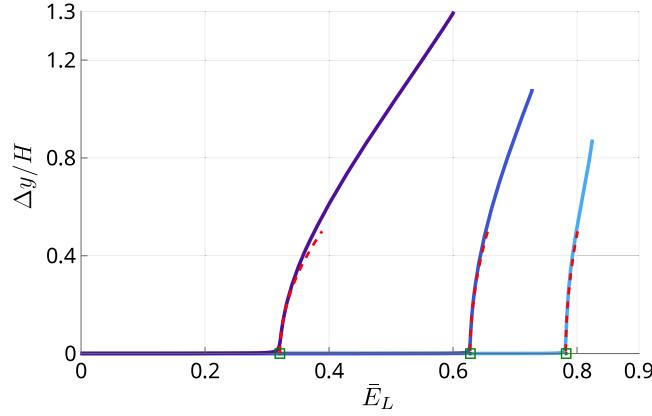
Recall that film-substrate systems may also exhibit period-doubling and period-tripling secondary bifurcations, see the works by Brau et al. (2011), Cao and Hutchinson (2012), Fu and Cai (2015), and Budday et al. (2015). For our soft dielectric film-substrate systems, we take a film 30 times stiffer than the substrate ( $r = 1/30$ ). To investigate secondary bifurcations, we conduct finite element simulations in computational domains that are twice and three times the fundamental length, respectively, while superposing imperfections corresponding to double and triple the critical wavelength. An intriguing feature of the system is the emergence of



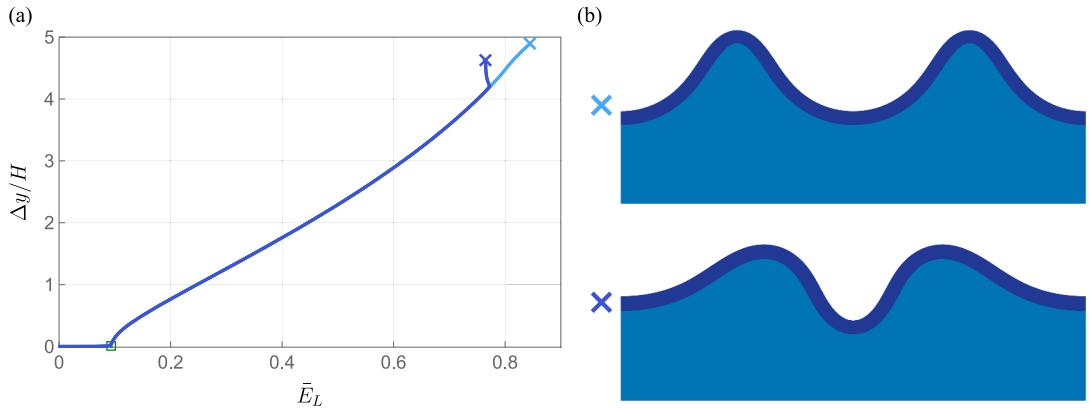
**Fig. 9.** (a) Plot of the non-dimensional amplitude of the wrinkling of the free surface,  $\Delta y/H$ , against the stretch  $\lambda$  for  $r = 1/5$  and  $\bar{E}_0 = 0.3$  (light blue line), 0.6 (blue line), and 0.75 (purple line). The green square denotes the marginal stability threshold obtained from the linearized stability analysis, while the cross indicates the onset of self-contact of the free surface. Letters mark the positions on the bifurcation diagram corresponding to the configurations shown below. The dashed lines show the best fit of the finite element data close to the bifurcation point with the function  $\hat{A}\sqrt{\lambda_{\text{cr}}^{\text{num}} - \lambda}$ , with  $\hat{A}, \lambda_{\text{cr}}^{\text{num}} > 0$ , see the main text for details. Here, the fitted parameter  $\hat{A} = 6.0952, 5.7925, 5.5954$  and  $\lambda_{\text{cr}}^{\text{num}} = 0.8481, 0.8936, 0.9378$  for  $\bar{E}_0 = 0.3, 0.6, 0.75$ , respectively. (b) Deformed configurations corresponding to the points A, B, C indicated in the bifurcation diagram, where the maximum principal stretch  $\lambda_{\max}$  (i.e., the square root of the maximum eigenvalue of  $\mathbf{F}^T \mathbf{F}$  for each point of the domain) and the non-dimensional electric potential field,  $\bar{\varphi} = \sqrt{\varepsilon/\mu_f}(\varphi/H)$ , are reported. A progressive strain localization is observed in the furrows of the wrinkling pattern beyond the turning point B, eventually leading to crease formation in the point C.



**Fig. 10.** Results of the finite element simulations for  $r = 1/30$  and  $\bar{E}_0 = 0.3$ . (a) Non-dimensional amplitude of the wrinkling of the free surface  $\Delta y/H$  versus the stretch  $\lambda$ . The light blue line represents the wrinkling solution with a constant wavelength, while the blue and purple lines show the amplitude of the wrinkling pattern when period-doubling and period-tripling secondary bifurcations occur. The green square denotes the marginal stability threshold predicted by the linearized stability analysis, while the cross indicates when self-contact of the film occurs. The fitting of Eq. (21) is not shown here, as its range of validity is too limited compared to the large amplitude of the wrinkling pattern. (b) Final morphologies of the finite element simulations at the onset of self-contact (represented in the inset), for the fixed-wavelength, period-doubling, and period-tripling solutions.



**Fig. 11.** Bifurcation diagrams showing the non-dimensional wrinkling amplitude  $\Delta y/H$  versus the applied non-dimensional voltage  $\bar{E}_L$  for  $r = 1/5$  and  $\lambda = 0.85, 0.9$ , and  $0.95$  (purple, blue, and light blue lines, respectively). The green squares denote the marginal stability thresholds obtained from the linearized stability analysis. The dashed lines show the best fit of the finite element data close to the bifurcation point with the function  $\hat{A}\sqrt{\bar{E}_L - \bar{E}_{\text{num}}^{\text{cr}}}$  with  $\hat{A} > 0$ , see the main text for details. Here, the fitted parameter  $\hat{A} = 1.9198, 2.9683, 3.6174$  and  $\bar{E}_{\text{num}}^{\text{cr}} = 0.3214, 0.6278, 0.7819$  for  $\lambda = 0.85, 0.9, 0.95$ , respectively.



**Fig. 12.** (a) Bifurcation diagrams showing the non-dimensional wrinkling amplitude  $\Delta y/H$  versus the applied non-dimensional voltage  $\bar{E}_L$  for  $r = 1/30$  and  $\lambda = 0.95$ . The green square denotes the marginal stability threshold obtained from the linearized stability analysis. The light blue line represents the wrinkling solution with a constant wavelength, while the blue line corresponds to the amplitude of the pattern after a period-doubling bifurcation occurs. The fitting of Eq. (21) is not explicitly shown here, as its range of validity is too limited compared to the large amplitude of the wrinkling pattern. (b) Final morphology from the finite element simulations corresponding to the end points of the two branches shown in Panel (a), for the fixed-wavelength solution (top figure, corresponding to the end point of the light blue branch) and the period-doubling solution (bottom figure, corresponding to the end point of the dark blue branch).

secondary bifurcations in the form of period-doubling and period-tripling (see Fig. 10), whose occurrence is strongly influenced by the selected computational domain size and the characteristic length of the imposed imperfections. Again, we note turning points in the nonlinear regime. In these cases as well, the turning points correspond to a localization of the deformation close to the wrinkling furrows, which later evolve into self-contacting creases.

We also analyze the behavior of the system when  $\lambda$  is held fixed and the applied non-dimensional voltage  $\bar{E}_L$  is used to trigger the wrinkling instability. It is observed from Fig. 11 that the system undergoes a supercritical transition at the onset of instability. In contrast to the stretch-induced case, the finite element simulations in this scenario terminate before the onset of strain localization. The interruption of some of the bifurcation branches in Fig. 11 arises from this loss of convergence. The arclength algorithm halves the step size whenever Newton's method fails to converge; a branch is therefore terminated once this halving is performed ten consecutive times, indicating significant numerical difficulty. Several factors may contribute to this behavior. One is the onset of strain localization associated with the catastrophic thinning of the soft dielectric layer (Zurlo et al., 2017), a phenomenon associated with the loss of convexity in the energy functional, potentially leading to the non-existence of energy minimizers. Another potential source of numerical difficulties is volumetric locking due to the incompressibility constraint. While advanced mixed formulations, such as the Hu-Washizu approach (Bonet and Wood, 1997), can alleviate locking in standard nonlinear elasticity, the situation is more complex in nonlinear electro-elasticity because of its multi-physical nature. Although enhanced formulations tailored for electro-mechanical coupling have been proposed (Gil and Ortigosa, 2016), their implementation lies beyond the scope of the present work.

As in the previous case, the bifurcation curves closely resemble those of supercritical pitchfork transitions. To verify this observation, we fit the finite element numerical data using the function  $\hat{A}\sqrt{|\bar{E}_L - \bar{E}_{\text{num}}^{\text{cr}}|}$ , following the same fitting procedure described earlier. The bifurcation is considered supercritical if  $\bar{E}_L > \bar{E}_{\text{num}}^{\text{cr}}$  close to the bifurcation point, and otherwise subcritical. The agreement is excellent near the bifurcation point, with the numerically predicted thresholds  $\bar{E}_{\text{num}}^{\text{cr}}$  deviating from the theoretical values by less than 1% in almost all cases, see Fig. 11 and Table 2 for a quantitative comparison with the theoretical thresholds. For the case  $r = 1/5$  and  $\lambda = 1.2$ , the finite element simulations are highly sensitive to the imposed imperfection. To trigger the instability, we have to increase the imperfection amplitude to  $1.5 \times 10^{-4}$  (compared to the baseline value of  $5 \times 10^{-5}$ , with all lengths scaled by the coating thickness). This adjustment causes the finite element numerical prediction of the critical voltage to appear at a lower value. Although decreasing the imperfection amplitude improves the accuracy of the critical threshold, in this case the simulations fail to converge once the bifurcation point is reached.

Furthermore, we observe period-doubling bifurcations when the soft dielectric film is sufficiently stiff relative to the substrate ( $r = 1/30$ ), as illustrated in Fig. 12(a). In the bifurcation diagram, we find excellent agreement with the linearized stability analysis. Similarly to the simulations shown in Fig. 11, we do not observe any strain localization or self-contact. Instead, the resulting post-bifurcation morphology features ridges separated by elongated furrows, as displayed in Fig. 12(b). Interestingly, a ridge morphology emerges here as a result of the instability for relatively large values of  $r$  compared to purely elastic passive systems, where  $r < 10^{-3}$ , see Wang and Zhao (2015).

#### 4. Conclusions

We presented a comprehensive theoretical analysis of the wrinkling instability of a soft dielectric film bonded to a hyperelastic substrate under the combined action of applied voltage and plane-strain mechanical loading.

By relying on the Stroh formulation and the surface impedance matrix method, we obtained exact bifurcation equations and accurate sixth-order approximate bifurcation equations. We also derived explicit bifurcation equations of critical stretch  $\lambda_{\text{cr}}$ , voltage  $\bar{E}_{L\text{cr}}$  and wavenumber  $(kh)_{\text{cr}}$ . The asymptotic solution agrees well with the exact solution when  $r$  is small, meeting the assumptions (of order  $(kh)^3$  for small  $kh$ ). Furthermore, we found that the thresholds of the shear modulus ratio  $r_c^0$  and pre-stretch  $\lambda_c^0$  for electro-elastic wrinkling correspond to the purely mechanical instability case.

Finally, our finite element simulations further enriched these findings by exploring post-buckling behavior and complex pattern evolution beyond the initial wrinkle formation. The simulations confirmed that the analytical critical points accurately mark the onset of instability, and revealed what happens beyond this point. We observed secondary bifurcations such as period-doubling and tripling of the wrinkle pattern when the film is relatively stiff compared to the substrate. These secondary patterns imply that a single system can support multiple modes of surface morphology, which could be harnessed to achieve different functional states (for example, switching between two distinct wrinkle wavelengths under different electrical inputs).

The simulations also uncovered a limit to the tunability, because at high voltage levels (beyond a turning point in the bifurcation diagram), localized strain concentrations can lead to the formation of a sharp crease with self-contact. This incipient creasing is a critical consideration for applications, as it represents a material failure or extreme deformation state that designers might wish to avoid.

Thus, our results not only map out the desired regime for reversible wrinkling but also delineate the boundaries where the surface topology might become unstable in a destructive way. For future work, we plan to experimentally validate the predicted electro-mechanical wrinkling and creasing instabilities. The theoretical analyses and finite element simulations presented here provide essential guidance for designing these experiments and ensure they build directly on the insights from our modeling.

#### CRediT authorship contribution statement

**Bin Wu:** Writing – review & editing, Writing – original draft, Validation, Supervision, Software, Methodology, Formal analysis, Data curation, Conceptualization; **Linghao Kong:** Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Conceptualization; **Weiqiu Chen:** Writing – review & editing, Supervision, Formal analysis, Conceptualization; **Davide Riccobelli:** Writing – review & editing, Visualization, Validation, Software, Methodology, Investigation, Formal analysis; **Michel Destrade:** Writing – review & editing, Methodology, Funding acquisition, Formal analysis, Conceptualization.

#### Data availability

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

#### 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.

During the preparation of this work, the authors used ChatGPT to correct grammatical mistakes in the original text, ensure that the paragraph structure and the language were clear and cohesive, and check some algebraic and asymptotic calculations. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the published article.

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## Appendix A. Exact and approximate bifurcation equations

The basic equations governing the finite electro-elastic deformations of an incompressible soft electro-elastic body are well-established, and there is no need to repeat them here. The same remark applies to the Stroh formulation of the equations of incremental deformations with sinusoidal variations along  $x_1$  and exponential variations along  $x_2$ . We refer the interested reader to the works of Dorfmann and Ogden (2014), Su et al. (2018), Dorfmann and Ogden (2019), Broderick et al. (2020), Su et al. (2020a) and Yang and Sharma (2023), for example.

As summarized in Section 2.2, the generalized, non-dimensional displacement-traction vector  $\eta$  satisfies  $\eta' = iN\eta$ , where  $i = \sqrt{-1}$  is the imaginary unit, the prime denotes differentiation with respect to  $kx_2$ , and  $N$  is the (constant) Stroh matrix, which is partitioned as  $N = \begin{bmatrix} N_1 & N_2 \\ N_3 & N_1^T \end{bmatrix}$ . For a general triaxial pre-stretch  $(\lambda_1, \lambda_2, \lambda_3)$ , we find that for the dielectric film characterized by the neo-Hookean ideal dielectric model (1),

$$N_1 = \begin{bmatrix} 0 & -1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad N_2 = \begin{bmatrix} \lambda_1^2 \lambda_3^2 & 0 & \lambda_1^3 \lambda_3^3 \bar{E}_L \\ 0 & 0 & 0 \\ \lambda_1^3 \lambda_3^3 \bar{E}_L & 0 & 1 + \lambda_1^4 \lambda_3^4 \bar{E}_L^2 \end{bmatrix},$$

$$N_3 = \begin{bmatrix} -(\lambda_1^2 + 3\lambda_1^{-2}\lambda_3^{-2} + 3\lambda_1^2\lambda_3^2\bar{E}_L^2) & 0 & 2\lambda_1\lambda_3\bar{E}_L \\ 0 & \lambda_1^{-2}\lambda_3^{-2} - \lambda_1^2 + \lambda_1^2\lambda_3^2\bar{E}_L^2 & 0 \\ 2\lambda_1\lambda_3\bar{E}_L & 0 & -1 \end{bmatrix}, \quad (A.1)$$

in our non-dimensional form, with  $\bar{E}_L = \sqrt{\epsilon/\mu_f}(V/H)$  representing the non-dimensional voltage (for more general formulas, see Su et al. (2018), where  $N$  is derived for a generic total free energy density function). The eigenvalues of  $N$  with positive imaginary parts are  $q_1 = i$ ,  $q_2 = i\lambda_1^2\lambda_3$ ,  $q_3 = i$ , and the corresponding eigenvectors are the columns of the  $6 \times 3$  matrix below,

$$[\eta^{(1)}|\eta^{(2)}|\eta^{(3)}] = \begin{bmatrix} -i\lambda_1^2\lambda_3^2 & -i\lambda_1^4\lambda_3^3 & 0 \\ \lambda_1^2\lambda_3^2 & \lambda_1^2\lambda_3^3 & 0 \\ -i\lambda_1^3\lambda_3^3\bar{E}_L & -i\lambda_1^5\lambda_3^3\bar{E}_L & i \\ 2 + \lambda_1^4\lambda_3^4\bar{E}_L^2 & 1 + \lambda_1^4\lambda_3^2 + \lambda_1^4\lambda_3^4\bar{E}_L^2 & \lambda_1\lambda_3\bar{E}_L \\ i(1 + \lambda_1^4\lambda_3^2) & 2i\lambda_1^2\lambda_3 & i\lambda_1\lambda_3\bar{E}_L \\ -\lambda_1^3\lambda_3^3\bar{E}_L & -\lambda_1^3\lambda_3^3\bar{E}_L & -1 \end{bmatrix}. \quad (A.2)$$

The eigenvalues with negative imaginary parts are  $q_4, q_5, q_6$  with associated eigenvectors  $\eta^{(4)}, \eta^{(5)}, \eta^{(6)}$ , which are the complex conjugates of  $q_1, q_2, q_3$  and  $\eta^{(1)}, \eta^{(2)}, \eta^{(3)}$ , respectively. The  $6 \times 6$  complete matrix of eigenvectors is defined as  $\mathcal{N} = [\eta^{(1)}|\eta^{(2)}|\eta^{(3)}|\eta^{(4)}|\eta^{(5)}|\eta^{(6)}]$ .

If the soft dielectric material were to occupy an entire half-space, then its impedance matrix would be  $Z = -iBA^{-1}$ , where  $A$  and  $B$  are the  $3 \times 3$  top and bottom submatrices of Eq. (A.2), respectively, or

$$Z = \begin{bmatrix} \lambda_1^{-2}\lambda_3^{-2} + \lambda_3^{-1} + \lambda_1^2\lambda_3^2\bar{E}_L^2 & -i(\lambda_1^{-2}\lambda_3^{-2} - \lambda_3^{-1} + \lambda_1^2\lambda_3^2\bar{E}_L^2) & -\lambda_1\lambda_3\bar{E}_L \\ i(\lambda_1^{-2}\lambda_3^{-2} - \lambda_3^{-1} + \lambda_1^2\lambda_3^2\bar{E}_L^2) & \lambda_1^2 + \lambda_3^{-1} & -i\lambda_1\lambda_3\bar{E}_L \\ -\lambda_1\lambda_3\bar{E}_L & i\lambda_1\lambda_3\bar{E}_L & 1 \end{bmatrix}. \quad (A.3)$$

The bifurcation condition for the Biot-type surface instability would then be:  $\det Z = 0$  (Destrade et al., 2008; Destrade, 2015), or

$$\lambda_1^6\lambda_3^3 + \lambda_1^4\lambda_3^2 + 3\lambda_1^2\lambda_3 - 1 = \lambda_1^4\lambda_3^4(1 + \lambda_1^2\lambda_3)\bar{E}_L^2. \quad (A.4)$$

In plane strain ( $\lambda_1 = \lambda, \lambda_3 = 1$ ), the bifurcation Eq. (A.4) reduces to Eq. (7), while in equi-biaxial strain ( $\lambda_1 = \lambda_3 = \lambda$ ), it recovers the formula established by Su et al. (2018).

Here, however, the soft dielectric film has a finite thickness and is in contact with the elastic substrate. The  $3 \times 1$  generalized, non-dimensional traction  $S = [S_{21}, S_{22}, \Phi]^T$  and displacement  $U = [U_1, U_2, \Delta]^T$  vectors on each side of the interface at  $x_2 = 0$  are related through

$$S_f(0) = iZ_f U_f(0), \quad S_s(0) = iZ_s U_s(0), \quad (A.5)$$

so that the boundary conditions of perfect bond at the interface,  $U_f(0) = U_s(0)$  and  $\mu_f S_f(0) = \mu_s S_s(0)$ , yield the bifurcation condition as Eq. (3):  $\det(Z_f - rZ_s) = 0$  (see, e.g., Shuvalov and Every (2002)). Here, the film impedance matrix  $Z_f$  at  $x_2 = 0$ , assuming the  $x_2 = -h$  surface is traction-free and the applied voltage remains constant, is defined as  $Z_f = -iM_3 M_1^{-1}$ , where  $M_1$  and  $M_3$  are, respectively,

the  $3 \times 3$  upper-diagonal and lower-off-diagonal submatrices of the  $6 \times 6$  exponential matrix  $\mathbf{M} = \exp(ikh\mathbf{N})$ , which can be computed as  $\mathbf{M} = \mathbf{N}\Lambda\mathbf{N}^{-1}$ , with  $\Lambda$  the diagonal matrix with elements  $e^{i\alpha_j kh}$  ( $j = 1, \dots, 6$ ). The substrate impedance matrix  $\mathbf{Z}_s$  reads as follows,

$$\mathbf{Z}_s = \begin{bmatrix} \lambda_1^{-2}\lambda_3^{-2} + \lambda_3^{-1} & -i(\lambda_1^{-2}\lambda_3^{-2} - \lambda_3^{-1}) & 0 \\ i(\lambda_1^{-2}\lambda_3^{-2} - \lambda_3^{-1}) & \lambda_1^2 + \lambda_3^{-1} & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad (\text{A.6})$$

which is consistent with Eq. (A.3) written at  $\bar{E}_L = 0$ , provided the last diagonal entry there is replaced with a zero to account for the two-dimensional nature of the traction and displacement vectors in the hyperelastic substrate (with no electric field).

We can solve the exact bifurcation Eq. (3) numerically, but it can prove computationally costly, which is why we may wish to use small-parameter expansions and conduct asymptotic analysis.

The solution of the first-order differential equation,  $\eta' = i\mathbf{N}\eta$ , with a constant Stroh matrix  $\mathbf{N}$ , is  $\eta(kx_2) = \exp(ikx_2\mathbf{N})\eta(0)$ . Therefore, the relationship between the generalized displacement-traction vectors of the upper and lower surfaces of the soft dielectric film reads  $\eta(-kh) = \exp(-ikh\mathbf{N})\eta(0) \equiv \hat{\mathbf{M}}\eta(0)$ . From the continuity conditions of  $\eta$  at the interface  $x_2 = 0$  ( $\mathbf{U}_f(0) = \mathbf{U}_s(0)$  and  $\mathbf{S}_f(0) = r\mathbf{S}_s(0)$ ), and the conditions of zero traction and a constant applied voltage on the top surface  $x_2 = -h$  ( $\mathbf{S}_f(-kh) = \mathbf{0}$ ), it follows that

$$\begin{bmatrix} \mathbf{U}_f(-kh) \\ \mathbf{0} \\ r\mathbf{S}_s(0) \end{bmatrix} = \hat{\mathbf{M}} \begin{bmatrix} \mathbf{U}_s(0) \\ \mathbf{0} \\ r\mathbf{S}_s(0) \end{bmatrix}. \quad (\text{A.7})$$

For thin dielectric films, where  $kh \ll 1$ , we substitute (A.5)<sub>2</sub> into (A.7) and write the power series  $\hat{\mathbf{M}} \equiv \exp(-ikh\mathbf{N}) = \sum \frac{1}{n!}(-i\mathbf{N})^n(kh)^n$  to arrive at the sixth-order approximation of the exact bifurcation equation,

$$\det \begin{bmatrix} ir\mathbf{Z}_s + (r\mathbf{N}_1\mathbf{Z}_s - i\mathbf{N}_3)(kh) - \frac{1}{2}i(r\mathbf{K}_4^{(2)}\mathbf{Z}_s - i\mathbf{K}_3^{(2)})(kh)^2 \\ -\frac{1}{6}(r\mathbf{K}_4^{(3)}\mathbf{Z}_s - i\mathbf{K}_3^{(3)})(kh)^3 + \frac{1}{24}i(r\mathbf{K}_4^{(4)}\mathbf{Z}_s - i\mathbf{K}_3^{(4)})(kh)^4 \\ +\frac{1}{120}(r\mathbf{K}_4^{(5)}\mathbf{Z}_s - i\mathbf{K}_3^{(5)})(kh)^5 - \frac{1}{720}i(r\mathbf{K}_4^{(6)}\mathbf{Z}_s - i\mathbf{K}_3^{(6)})(kh)^6 \end{bmatrix} = 0, \quad (\text{A.8})$$

where  $\mathbf{K}_3^{(n)}$  and  $\mathbf{K}_4^{(n)}$  are, respectively, the  $3 \times 3$  lower-off-diagonal and lower-diagonal submatrices of the  $6 \times 6$  matrix  $\mathbf{K}^{(n)} \equiv \mathbf{N}^n$ .

Solving this approximate bifurcation Eq. (A.8) numerically is much more efficient and less computationally expensive than solving the exact bifurcation condition (3), and it is highly accurate for small  $kh$  and small  $r$ .

## Appendix B. Asymptotic analysis of approximate bifurcation Eq. (A.8)

Following Cai and Fu (2000), we can use the approximate bifurcation Eq. (A.8) in this appendix to derive power-series asymptotic expansions in  $kh$  for the stretch  $\lambda$  and voltage  $\bar{E}_L$  when the soft dielectric film is much stiffer than the substrate, and further, explicit asymptotic expansions of the critical values  $\lambda_{\text{cr}}$  and  $\bar{E}_L^{\text{cr}}$  in powers of  $r^{1/3}$ . Here we focus on the plane-strain loading case.

Assuming  $kh \ll 1$  and  $r$  of order  $(kh)^3$ , an expansion of the sixth-order approximate bifurcation condition (A.8), followed by elimination of the common factor, leads to

$$\begin{aligned} \omega_0 + \omega_1(kh) + \frac{1}{2}\omega_2(kh)^2 + \frac{1}{6}\omega_3(kh)^3 + \frac{1}{24}\omega_4(kh)^4 \\ + \frac{1}{120}\omega_5(kh)^5 + \frac{1}{720}\omega_6(kh)^6 + \mathcal{O}((kh)^7) = 0, \end{aligned} \quad (\text{B.1})$$

where

$$\begin{aligned} \omega_0 &= (-1 + 3\lambda^2 + \lambda^4 + \lambda^6)r^2, \\ \omega_1 &= -(1 + \lambda^2)[1 - 3\lambda^2 + (\bar{E}_L^2 - 1)\lambda^4(1 + \lambda^2)]r, \\ \omega_2 &= 2(\bar{E}_L^2 - 1)[(\bar{E}_L^2 - 1)\lambda^4 - 2]\lambda^4 - 6 - 8(\lambda^2 - 1)r, \\ \omega_3 &= -(1 + \lambda^2)\{2 - 4\lambda^2 + \lambda^4[-7 - 2\bar{E}_L^4\lambda^4 - \lambda^2(7 + 3\lambda^2 + \lambda^4) \\ &\quad + \bar{E}_L^2(6 + 4\lambda^2 + 5\lambda^4 + \lambda^6)]\}r, \\ \omega_4 &= -16 + 4\lambda^4[5 - 2\bar{E}_L^2 + 2(3 - 4\bar{E}_L^2 + \bar{E}_L^4)\lambda^4 + (\bar{E}_L^2 - 1)^2\lambda^8], \\ \omega_5 &= f(\lambda, \bar{E}_L^2)r, \\ \omega_6 &= -40 - 8(\bar{E}_L^2 - 9)\lambda^4 + 2(91 - 92\bar{E}_L^2 + 16\bar{E}_L^4)\lambda^8 \\ &\quad + 4(17 - 27\bar{E}_L^2 + 10\bar{E}_L^4)\lambda^{12} + 6(\bar{E}_L^2 - 1)^2\lambda^{16}, \end{aligned} \quad (\text{B.2})$$

where we omit the explicit form of  $f(\lambda, \bar{E}_L^2)$  for brevity. Because  $\omega_5$  depends linearly on  $r$ , the sixth term in Eq. (B.1) is of order  $(kh)^8$  and may consequently be discarded.

### B.1. Critical stretch under a prescribed electric voltage

For a prescribed non-dimensional electric voltage  $\bar{E}_L = \bar{E}_0$ , we first derive a power-series asymptotic expansion of the stretch  $\lambda$  in  $kh$ , from which the explicit asymptotic expansion of the critical stretch  $\lambda_{\text{cr}}$  in powers of  $r^{1/3}$  follows.

As  $r = \mathcal{O}((kh)^3)$ , the leading-order term arises from the third term in Eq. (B.1), which is of order  $(kh)^2$ . Thus, Eq. (B.1) reduces to the leading-order bifurcation condition,

$$3 - (1 - \bar{E}_0^2)\lambda^4[2 + (1 - \bar{E}_0^2)\lambda^4] = 0, \quad (\text{B.3})$$

which gives the leading-order expression for the (critical) stretch,  $\lambda_0 = (1 - \bar{E}_0^2)^{-1/4}$ , as presented in Eq. (2).

Examination of Eq. (B.1) reveals that the coefficient  $\phi_1$  in the first-order asymptotic expansion  $\lambda = \lambda_0 + \phi_1 kh$  vanishes, and that the next-order expansion is  $\lambda = \lambda_0 + \phi_2(kh)^2$ . Substituting this into Eq. (B.1) and equating the coefficients of  $(kh)^4$  yields

$$1 + 3(1 + \lambda_0^{-2})r/(kh)^3 + 12\lambda_0^{-5}\phi_2 = 0, \quad (\text{B.4})$$

which gives  $\phi_2$ , and the second-order correction to the stretch as

$$\lambda = \lambda_0 - \frac{1}{4}(\lambda_0^3 + \lambda_0^5)\left(\frac{r}{kh}\right) - \frac{1}{12}\lambda_0^5(kh)^2. \quad (\text{B.5})$$

In a similar manner, substituting the third- and fourth-order asymptotic expansions,  $\lambda = \lambda_0 + \phi_2(kh)^2 + \phi_3(kh)^3$  and  $\lambda = \lambda_0 + \phi_2(kh)^2 + \phi_3(kh)^3 + \phi_4(kh)^4$ , into Eq. (B.1) and equating the coefficients of  $(kh)^5$  and  $(kh)^6$  yields  $\phi_3$  and  $\phi_4$ , respectively. Their explicit forms are omitted here for brevity. The resulting fourth-order asymptotic expansion of the stretch  $\lambda$  is presented in Eq. (13).

Subsequently, we determine, in turn, the critical wavenumber  $(kh)_{\text{cr}}$  in Eq. (14) and the critical stretch  $\lambda_{\text{cr}}$  in Eq. (15) by setting the derivative of Eq. (13) with respect to  $kh$  equal to zero.

## B.2. Critical electric voltage for a fixed pre-stretch

Here we derive a power-series asymptotic expansion in  $kh$  for  $\bar{E}_L^2$  at a fixed pre-stretch  $\lambda$ , from which the explicit asymptotic expansion of the squared critical voltage  $(\bar{E}_L^{\text{cr}})^2$  in powers of  $r^{1/3}$  can be obtained.

Analogous to the derivation of the asymptotic expansion of the stretch  $\lambda$  presented in Appendix B.1, the leading-order term of the squared voltage is obtained from Eq. (B.3) as  $\bar{E}_{L0}^2 = 1 - \lambda^{-4}$ . In a similar manner, the second-order asymptotic expansion of the squared voltage can be derived as

$$\bar{E}_L^2 = \bar{E}_{L0}^2 + \frac{1}{3}(kh)^2 + (1 + \lambda^{-2})(r/kh), \quad (\text{B.6})$$

and the resulting fourth-order asymptotic expansion of  $\bar{E}_L^2$  is formulated in Eq. (18).

By subsequently setting the derivative of Eq. (18) with respect to  $kh$  to zero, the asymptotic expansions of the critical wavenumber  $(kh)_{\text{cr}}$  and the critical squared voltage  $(\bar{E}_L^{\text{cr}})^2$  in powers of  $r^{1/3}$  are obtained, as given in Eqs. (20) and (19), respectively.

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