

# FLOQUET ANALYSIS FOR STABILITY OF ELECTROLYTE FILMS ON SUBSTRATES WITH STRONGLY NONUNIFORM CHARGE DENSITY

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*We consider steady-state shapes and stability of a liquid electrolyte film on a substrate that is patterned or structured, resulting in a spatially periodic variation of the charge density. The electrical field in the liquid is described using the Debye-Hückel approximation to the classical Poisson-Boltzmann equation. Thin-film approximation is used to model liquid flow arising as a result of the interfacial instability. Steady-state shapes are found numerically for a range of conditions and shown to agree with the asymptotic results for the weak substrate charge nonuniformity. Floquet analysis is used to investigate the stability of the spatially periodic steady-state solutions. Linear stability criteria are found for a range of conditions. Depending on the wavelength of the substrate patterning, the effect can be either stabilizing or destabilizing, with the latter typically observed for smaller pattern wavelengths.*

**KEY WORDS:** electrolyte, liquid flow, electrical double layer

## 1. INTRODUCTION

Stability of thin liquid layers has been studied for many decades, as discussed in the well-known review articles of Oron et al. (1997) and Craster and Matar (2009). Several instability mechanisms have been investigated experimentally and described theoretically. While many of these mechanisms are relevant for the films of liquids containing ions, the key issue for stability of such films is the effect of electrical charges and fields. Mathematical modeling of electrolyte films has been an increasingly active area of research in recent years. Conroy et al. (2010) conducted a pioneering study of instability of electrolyte films. Their model was based on the assumption of the film thickness being much smaller than the relevant length scales along the fluid interface, leading to a lubrication-type formulation, widely used in the past for a related problem of dynamics of the so-called leaky dielectric films (Saville, 1997; Papageorgiou and Petropoulos, 2004). The effects of electrical charges and fields, surface tension, and viscous flow were incorporated into the formulation. The geometric configuration considered was that of core-annular flow in which an annular electrolyte film surrounds a perfectly conducting fluid core inside a cylindrical tube. Conditions for film rupture were identified and new strongly nonlinear steady-state solutions were found in the numerical simulations.

Ketelaar and Ajaev (2014a) investigated stability and nonlinear evolution of thin electrolyte liquid films on flat charged substrates. The effect of charge regulation at the solid substrate was incorporated into the model via a linear relation between the charge density and electric potential. The liquid-gas interface was assumed to be uniformly charged. Stability conditions were obtained for a range of values of film thickness and electrolyte concentration. The electric field in the liquid was found from the Debye-Hückel approximation to the Poisson-Boltzmann equation. A follow-up study of Ketelaar and Ajaev (2014b) addressed the same issues using the full nonlinear Poisson-Boltzmann equation for a symmetric electrolyte to describe the electrical field in the liquid. Instability of thin electrolyte films in the presence of tangential electrical field was studied by Ganchenko et al. (2015) while the effects of external time

### NOMENCLATURE

$d$	average liquid film thickness	$\gamma$	perturbation growth rate
$E, F$	matrices in Floquet analysis	$\delta$	charge nonuniformity parameter, see Eq. (1)
$h$	nondimensional film thickness	$\varepsilon$	dielectric permittivity
$k$	imaginary part of Floquet exponent	$\zeta$	interface perturbation amplitude
$L$	nondimensional length of the interval of periodicity	$\zeta_n$	Fourier coefficients of $\zeta$
$L_x$	dimensional length scale in the horizontal direction	$\kappa$	ratio of average film thickness to the Debye length
$p$	scaled pressure	$\lambda_D$	Debye length for the liquid electrolyte
$\tilde{q}$	scaled substrate charge density	$\mu$	viscosity
$\hat{q}$	scaled liquid surface charge density	$\rho$	density
$q_a$	average substrate charge density	$\sigma$	surface tension
$t$	nondimensional time	$\psi$	scaled electric potential
$U$	characteristic liquid velocity	$\hat{\psi}$	scaled potential at the fluid interface
$u$	scaled velocity along substrate	<b>Subscripts</b>	
$v$	scaled vertical velocity	0	steady-state
$x, y$	nondimensional Cartesian coordinates	$c$	critical

periodic electrical field in the direction normal to the fluid interface were considered by Bandopadhyay and Hardt (2017) in a configuration with two fluid layers.

Studies of electrolyte films are important for applications in which electrical fields are used to manipulate liquids, particles, and biological cells, e.g., in microfluidics, but these studies are also relevant for heat transfer applications since water used, e.g., in cooling systems, typically contains ions while interfaces are likely to carry some electrical charge. Liquid film rupture conditions are important in this context since the appearance of dry patches leads to a significant reduction of efficiency of heat transfer, as discussed, e.g., by Kabov (2000) and Zaitsev et al. (2005, 2007).

Models of electrolyte films typically assume that the substrate charge density is uniform. However, in applications, the interfacial charge density can be spatially nonuniform, especially in biological systems. This motivated substantial experimental work on the topic using artificially created surfaces with spatial charge density variation. For example, Silbert et al. (2012) created a heterogeneously charged substrate with domains of positively charged surfactant on mica substrate; bare mica is negatively charged. The outcome of these experimental works was a stronger than expected interaction between heterogeneously charged surfaces. Spatially periodic variation of charge density is a feature of many structured substrates, as discussed, e.g., in Squires (2008), Bahga et al. (2010), and Ajaev et al. (2011, 2016). Despite substantial interest in applications involving heterogeneous substrates, the topic of stability of electrolyte films on such substrates has received little attention. This is in contrast to situations involving nonelectrolyte films on heterogeneous substrates, which were analyzed using both numerical techniques and Floquet analysis, the latter being applied to cases of periodic variation of substrate properties. Examples of such situations are gravity-driven liquid films on inclined corrugated surfaces studied, e.g., by Dávalos-Orozco (2007), Tseluiko et al. (2013), and Trifonov (2014).

In our previous study, we considered a thin electrolyte film on a spatially nonuniform substrate (Jutley and Ajaev, 2018). It was found that depending on the parameter values the effect of the substrate charge nonuniformity can be either stabilizing or destabilizing. In addition to stability criteria, pattern formation in the strongly nonlinear regime was investigated. However, this work has two important limitations. First, the study was focused on relatively small departures of the charge density from its average value. Second, the results presented in the paper only included sinusoidal variation of the charge density. In the present study, we overcome both of these limitations.

## 2. FORMULATION

We consider a thin liquid layer of average thickness  $d$ , shown in Fig. 1, on a solid substrate with spatially periodic variation of the charge density. The presence of electrical charges at both liquid-solid and liquid-gas interfaces leads to formation of electrical double layers. As these layers overlap, spontaneous breakup of the liquid film is possible, as discussed, e.g., in Ketelaar and Ajaev (2014a). We address the conditions of such breakup for a situation when the solid substrate charge density (scaled by its average value  $q_a$ ) is written as

$$\tilde{q}(x) = 1 + \delta \sum_{n=0}^N \frac{(-1)^n}{2n+1} \cos \frac{(2n+1)\pi x}{L} \tag{1}$$

where  $x$  is the nondimensional coordinate in the direction along the substrate scaled by the quantity  $L_x$  defined below. For large  $N$ , this equation describes a square-wave spatial modulation of the substrate charge.

As the liquid film becomes unstable, its surface undergoes a deformation and viscous flow is generated in the film. Suppose the characteristic horizontal velocity and length scale for this flow are  $U$  and  $L_x$ , respectively. The balance between viscous and Maxwell stresses in the momentum equation leads to the condition

$$\frac{q_a^2}{\epsilon L_x} \sim \frac{\mu U}{d^2} \tag{2}$$

The value of  $L_x$  can be determined from the interfacial stress condition which incorporates both surface tension and electrostatic contribution, assumed to be of the same order so that

$$\frac{\sigma d}{L_x^2} \sim \frac{q_a^2}{\epsilon} \tag{3}$$

leading to  $L_x = \sqrt{\epsilon \sigma d} / q_a$ . Substituting this value into Eq. (2) gives the velocity scale,

$$U = \frac{q_a^3}{\mu \sigma^{1/2}} \left( \frac{d}{\epsilon} \right)^{3/2} \tag{4}$$

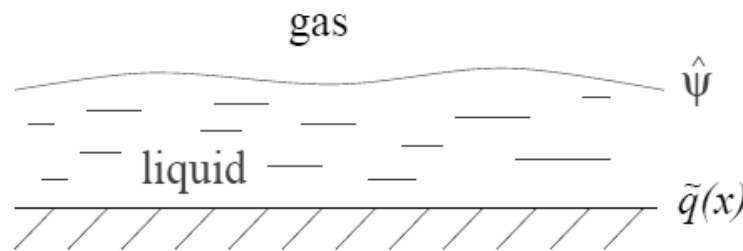
With the Cartesian coordinates  $x$  and  $y$  scaled by  $L_x$  and  $d$ , respectively, the governing equations become

$$p_x = u_{yy} + \psi \psi_x, \tag{5}$$

$$p_y = \psi \psi_y, \tag{6}$$

$$u_x + v_y = 0, \tag{7}$$

$$\psi_{yy} = \kappa^2 \psi, \quad \kappa = \frac{d}{\lambda_D}. \tag{8}$$



**FIG. 1:** Geometric configuration considered in our stability analysis. Both liquid-solid and liquid-gas interfaces carry interfacial charges.

Here the pressure is nondimensionalized by  $q_a^2/\varepsilon$ , the potential  $\psi$  by  $q_a\lambda_D/\varepsilon$ , and the vertical component of velocity  $v$  by  $dU/L_x$ . The governing equations are supplemented by the usual kinematic and stress conditions at the deformable interface, as well as the condition of zero velocity and given charge density at the substrate.

Equation (5) can be integrated twice, leading to a parabolic velocity profile. Once the velocity profile is established, the usual steps of the lubrication-type analysis (Oron et al., 1997; Craster and Matar, 2009) lead to the equation for film thickness of the form

$$h_t + [h^3(h_{xxx} - \hat{q}\hat{q}_x)]_x = 0. \quad (9)$$

Here the interfacial charge density  $\hat{q}$  is found from the solution of Eq. (8) with the substrate charge density given by (1) and interfacial potential  $\hat{\psi}$  assumed fixed. This solution can be written in explicit form:

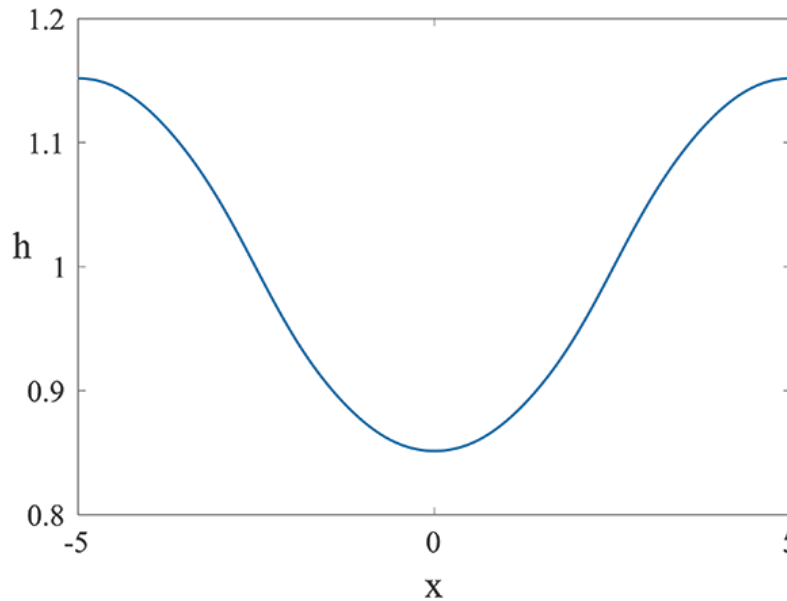
$$\hat{q} = \hat{\psi} \tanh(\kappa h) - \tilde{q} \operatorname{sech}(\kappa h). \quad (10)$$

### 3. STEADY-STATE SHAPES

Let us start with the determination of steady-state solutions of Eq. (9). Finite-difference discretization is used to reduce the solution of this equation to a linear system of differential equations for time-dependent values of the variable  $h$  on the spatially uniform grid. This system is then solved using a standard Matlab subroutine, `ode15s`, to obtain the time-dependent solution. The solution is monitored until the temporal change of its norm becomes less than a specified small value. The resulting profile is then considered to be the steady-state solution. A typical result is shown in Fig. 2. Note that the substrate charge variation is represented by a truncated Fourier series that simulates the square-wave variation, but the interface shape is nearly sinusoidal. Thus, the higher harmonics from the formula for the charge density are suppressed in the solution for the interface shape.

To better understand and verify these results, let us consider an approximate solution in the limit of small charge density variation amplitude  $\delta$ , i.e., consider an asymptotic expansion

$$h_0 = 1 + \delta h_0^{(1)} + \delta^2 h_0^{(2)} + \dots. \quad (11)$$



**FIG. 2:** Interface shape for  $\kappa = 1$ ,  $\hat{\psi} = -1$ ,  $L = 5$ ,  $\delta = 0.05$ ,  $N = 3$

This expansion can be substituted into Eq. (9), but it is more convenient to substitute it into the condition of constant pressure in the liquid, always satisfied for steady-state solutions and written in the form

$$h_{0xx} - \frac{1}{2}\hat{q}^2 = C. \tag{12}$$

After substituting the above asymptotic expansion for  $h_0$  into Eq. (12) and neglecting the  $O(\delta^2)$  terms, we obtain the following equation for  $h_0^{(1)}$ :

$$h_{0xx}^{(1)} - Gh_0^{(1)} = \tilde{G} \sum_{n=0}^N \frac{(-1)^n}{2n+1} \cos \frac{(2n+1)\pi x}{L}, \tag{13}$$

where

$$G = \hat{q} \frac{\partial \hat{q}}{\partial h} \Big|_{h=1}, \quad \tilde{G} = \hat{q} \frac{\partial \hat{q}}{\partial \tilde{q}} \Big|_{h=1}. \tag{14}$$

These quantities can be calculated from the explicit formula for  $\hat{q}$ , Eq. (10), leading to the solution of the differential equation for  $h_0^{(1)}$  in the form

$$h_0^{(1)} = \frac{\hat{\psi} \sinh \kappa - 1}{\cosh^2 \kappa} \sum_{n=0}^N \frac{(-1)^n}{2n+1} \left[ G + \frac{(2n+1)^2 \pi^2}{L^2} \right]^{-1} \cos \frac{(2n+1)\pi x}{L}. \tag{15}$$

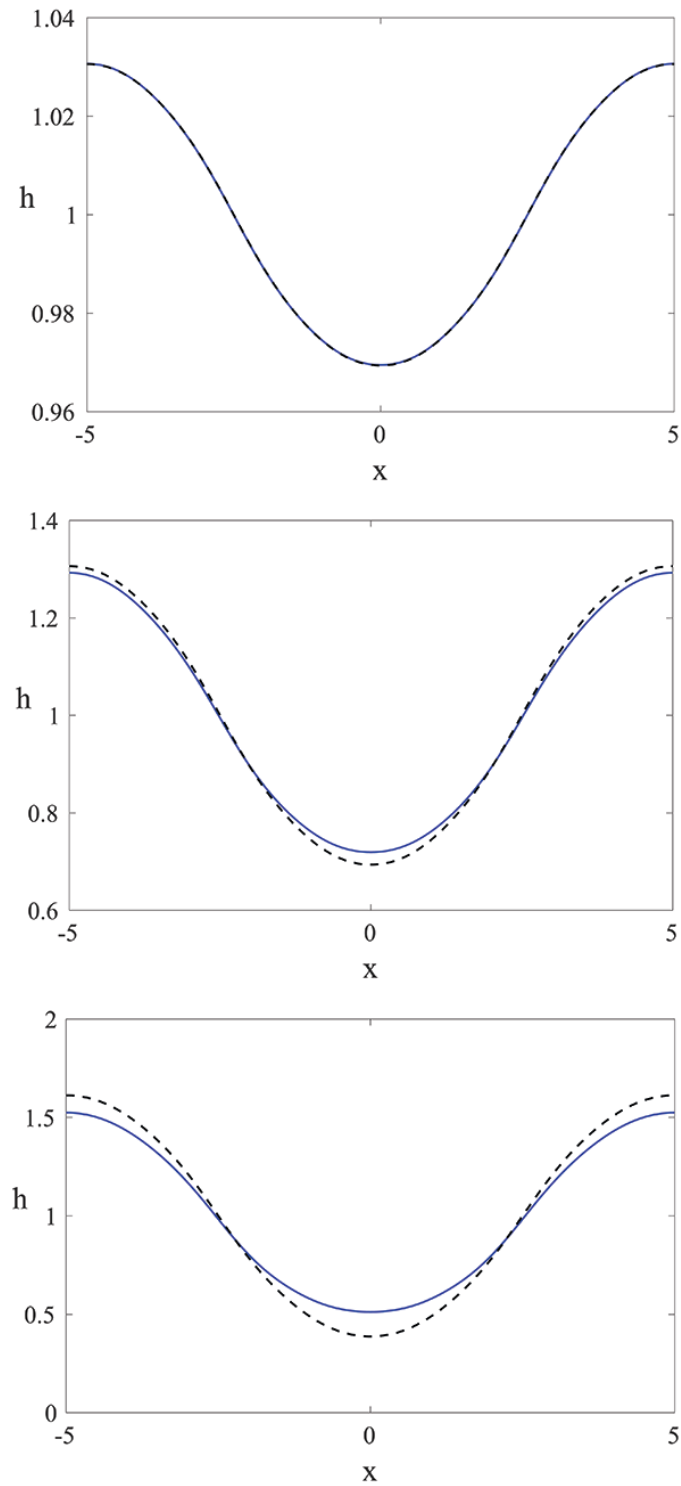
This equation clearly shows that the coefficients in the series expansion for  $h_0^{(1)}$  decay rapidly with increase of  $n$ . In fact, for small  $G$  the decrease is described by  $\sim (2n+1)^{-3}$ , significantly faster than the  $\sim (2n+1)^{-1}$  decay of the expansion coefficients of the function  $\tilde{q}(x)$ . This explains the fact that the solution above in Fig. 2 is close to sinusoidal even when the function  $\tilde{q}$  is basically a square wave. In physical terms, one can argue that the surface tension effects, represented by the second derivatives of the layer thickness, act to smooth the interface profile even in the regions where the underlying substrate properties are characterized by large spatial gradients.

Comparison of steady-state shapes between the approximate analytical solutions and the numerical results is illustrated in Fig. 3. The two are essentially identical for small  $\delta$ , but discrepancies are seen as  $\delta$  is increased. The approximate solution tends to overestimate the amplitude of the interface deformation.

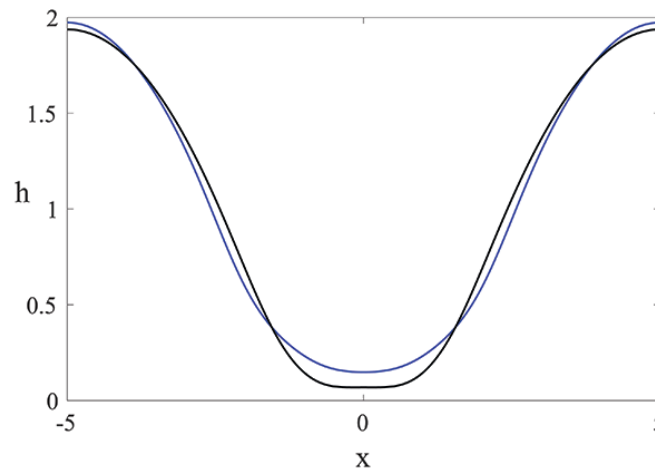
We have carried out detailed studies of the interface shapes for different parameters. Increasing  $\delta$  results in larger deformation amplitudes, which is clear from the results seen in Fig. 4. Note that the blue line corresponds to the same parameter values as in Fig. 2 except that  $\delta$  is increased to 0.5. In this regime, corresponding to strong deviations of the charge density from its average value, the small-amplitude approximation is no longer valid and the nonlinearity results in slight flattening of the interface. Both the amplitude and the flattening effect become more significant as the value of  $\kappa$  is increased, as seen from comparison of the two curves in Fig. 4. Larger  $\kappa$  corresponds to smaller  $\lambda_D$ , i.e., higher ion concentration in the liquid. Thus, it is expected that the electrostatic effects become more pronounced at these conditions.

#### 4. STABILITY ANALYSIS

The steady-state solutions discussed in the previous section are spatially periodic, so that to find them numerically it was sufficient to consider an interval of length  $2L$  rather than a much larger interval of the length  $2L_T$ , which is the horizontal extent of the entire patterned substrate, typically orders of magnitude larger than  $L$  in the configurations relevant to practical applications of patterned and structured substrates. For the stability problem, the wavelength of the perturbations can be expected to be comparable to  $L_T$  rather than  $L$ , so it may seem that computations on the large domain are required. However, similar reduction of the problem size can be achieved with the help of the classical Floquet theory which allows one to reduce the stability calculation to numerically finding a function  $\zeta(x)$  on the interval  $[-L, L]$ . The interface shape perturbation is then written as a superposition of terms of the form



**FIG. 3:** Interface shapes from the numerical solution (solid lines) and asymptotic analysis (dashed line) for  $\kappa = 1$ ,  $\hat{\psi} = -1$ ,  $L = 5$ ,  $N = 3$ , and three different values of  $\delta$ :  $\delta = 0.01$  (top),  $\delta = 0.1$  (middle), and  $\delta = 0.2$  (bottom)



**FIG. 4:** Steady-state interface shapes for  $\hat{\psi} = -1$ ,  $L = 5$ ,  $N = 3$ ,  $\delta = 0.5$ , with  $\kappa = 1$  (top line) and  $\kappa = 3$  (black line, flattened at the bottom)

$\zeta e^{\gamma t + ikx}$ . The linearized equation for this perturbation, derived from the general nonlinear equation (9) then leads to the following equation for  $\zeta$ :

$$h_0^3 D_k^4 \zeta + 3h_0^2 h_{0x} D_k^3 \zeta - h_0^3 Q D_k^2 \zeta - [2h_0^3 Q_x + 3h_0^2 h_{0xx} Q] D_k \zeta + (\gamma - 3h_0^2 h_{0xx} Q_x) \zeta = 0. \tag{16}$$

Here we introduce a new operator  $D_k$  and a new quantity  $Q$  defined by

$$D_k = \frac{\partial}{\partial x} + ik, \quad Q \equiv \hat{q}(h_0) \hat{q}_h(h_0). \tag{17}$$

Let us now represent the unknown periodic function  $\zeta$  by a Fourier series,

$$\zeta = \sum_{n=-N}^{N-1} \zeta_n e^{i\pi n x / L}. \tag{18}$$

Substitution into the equation above leads to a generalized eigenvalue problem,

$$F\mathbf{z} = \gamma E\mathbf{z}, \tag{19}$$

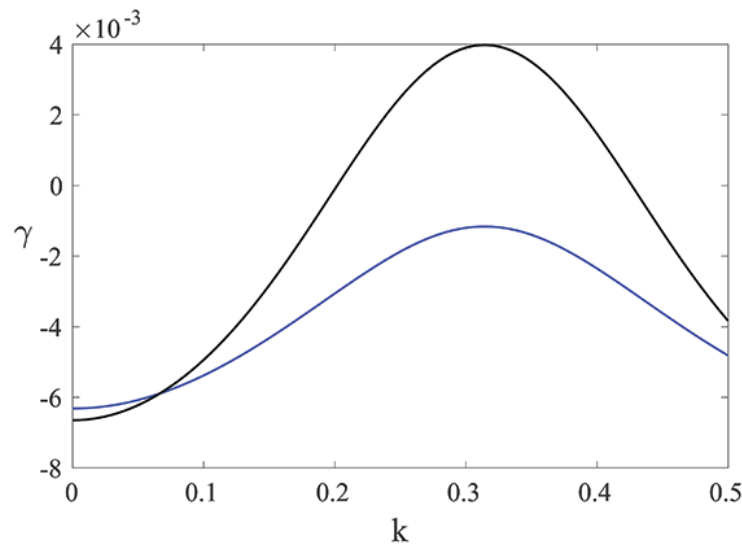
where  $\mathbf{z}$  is a vector with components equal to the Fourier coefficients  $\zeta_n$  and the matrices  $E$  and  $F$  are obtained from the discretized version of Eq. (16). Let us discuss their definition in more detail. It is convenient to introduce a summation index  $p$  such that  $p = n + 1 + N$ , which together with the summation index over the spatial grid,  $m$ , leads to the formulas

$$E_{mp} = e^{i\pi n x_m / L}, \tag{20}$$

$$F_{mp} = E_{mp} \left[ -h_{0m}^3 \left(k + \frac{\pi n}{L}\right)^4 + 3ih_{0m}^2 h_{0xm} \left(k + \frac{\pi n}{L}\right)^3 - h_{0m}^3 Q_m \left(k + \frac{\pi n}{L}\right)^2 + i(2h_{0m}^3 Q_{xm} + 3h_{0m}^2 h_{0xxm} Q_m) \left(k + \frac{\pi n}{L}\right) + h_{0m}^2 h_{0xxm} Q_{xm} \right], \tag{21}$$

where the subscript  $m$  denotes the values at the grid point number  $m$ .

Solving the generalized eigenvalue problem, (19), gives the value of the growth rate  $\gamma$  as a function of the magnitude  $k$  of the Floquet exponent, i.e., essentially the wave number. A typical result is shown in Fig. 5. We



**FIG. 5:** Dispersion curves obtained from Floquet analysis for  $\kappa = 1$ ,  $L = 5$ ,  $\delta = 0.05$ ,  $N = 3$ , and the scaled potential values of  $\hat{\psi} = -1.1$  (bottom line, smaller maximum value) and  $\hat{\psi} = -1.0$  (black line, larger maximum)

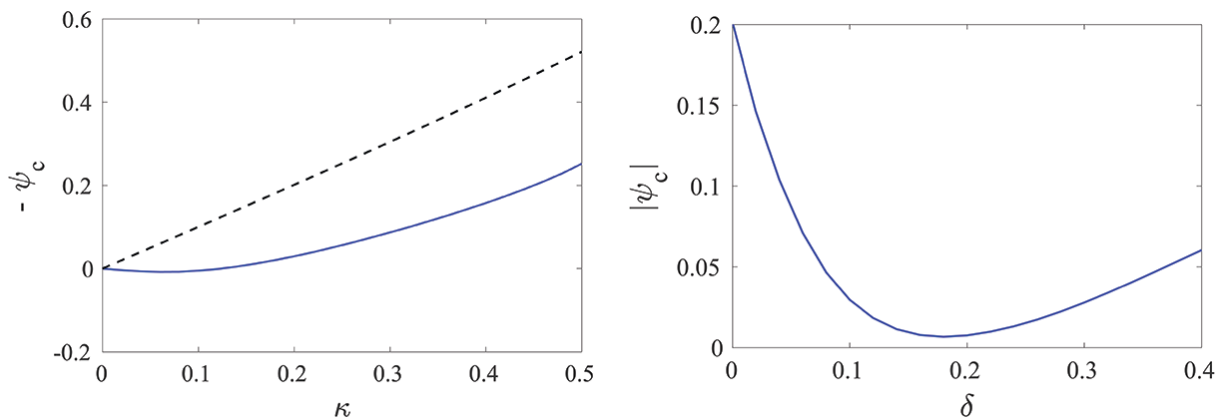
observe that the instability threshold for the film on uniform substrate corresponds to  $\psi_c = -1.1752$ , as discussed, e.g., by Jutley and Ajaev (2018). The lower absolute values of the potential correspond to unstable films. However, the negative values of growth rate with patterning for the blue line (the lower curve) in Fig. 5 indicate that the system is more stable. Only by decreasing the magnitude of potential further one observes transition to instability, i.e., positive growth rate for some of the disturbances, as illustrated by the black line (the curve with higher maximum) in the figure.

Stability criteria for a uniformly charged substrate can be obtained analytically, as discussed by, e.g., Ketelaar and Ajaev (2014a). For the case of  $\tilde{q} = 1$  and negative  $\hat{\psi}$ , the analytical approach leads to the critical value of  $\psi_c = -\sinh(\kappa)$ . We determined the shift in the value of  $\psi_c$  due to spatial variation of the charge density using the Floquet analysis discussed above. For each  $\kappa$ , the numerical solution of the generalized eigenvalue problem from (19) leads to determination of  $\gamma(k)$  and thus the maximum value of  $\gamma$ , denoted by  $\gamma_{\max}$ . Note that the shapes of the  $\gamma$  vs.  $k$  curves are qualitatively similar to the ones shown in Fig. 5. For different values of  $\hat{\psi}$ ,  $\gamma_{\max}$  will be different. The critical value  $\psi_c$  is defined by the condition of  $\gamma_{\max} = 0$ , which is determined numerically using the standard Matlab subroutine `fzero`.

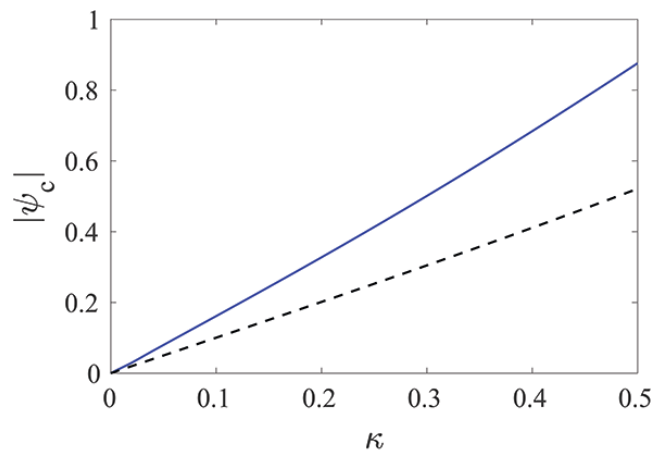
Figure 6 (left) shows the stability threshold, expressed in terms of  $-\psi_c$ , as a function of  $\kappa$ . The corresponding result for the uniformly charged substrate is shown by the dashed line for comparison. The charge nonuniformity has a stabilizing effect. Based on the results of Jutley and Ajaev (2018), obtained at small  $\delta$ , one might expect the stabilizing effect to be more significant for larger  $\delta$ . To test this hypothesis, we plot the stability threshold as a function of  $\delta$  in Fig. 6 (right). Surprisingly, the correction starts to decrease as  $\delta$  is increased above  $\sim 0.2$ . Thus, using the full nonlinear expression for the interface deformation results in a smaller value of the shift in the stability criterion than may be expected from the linearization in  $\delta$ . This is consistent with the fact that the nonlinear interface deformation seen in Fig. 3 is smaller than predicted by the asymptotic analysis valid for small  $\delta$ .

It has been suggested based on analytical considerations by Jutley and Ajaev (2018) that decreasing  $L$  can result in destabilization of the interface. This was verified previously by numerical solution obtained at small  $\delta$ . We investigated this further and found that destabilization for a wide range of parameters. A typical result is illustrated in Fig. 7. Note that the value of  $L$  is now reduced to  $L = 2$ . The dashed line still shows the stability boundary for uniformly charged substrate, with the unstable region below the curve. The solid line comes from the numerical stability analysis based on Floquet theory and shows significant expansion of the region of the instability. The effect is more pronounced as the value of  $\kappa$  is increased.





**FIG. 6:** Stability results for  $L = 5$ ,  $N = 3$ : (left) fixed  $\delta = 0.1$ , with the dashed line indicating the results at  $\delta = 0$ ; (right) fixed  $\kappa = 0.2$



**FIG. 7:** Stability curves for  $L = 2$ ,  $\delta = 0.1$  (solid line) and  $\delta = 0$  (dashed line),  $N = 3$

### 5. CONCLUSIONS

We use a lubrication-type approach to investigate steady states and stability for a thin electrolyte film on a charged substrate. Description of viscous flow in the liquid film is coupled to the solution of the equations for the electric potential in the limit of small potentials, i.e., in the Debye-Hückel approximation. The substrate charge density profile is chosen to simulate a square-wave profile, as appropriate for patterned and structured substrates. We found that the steady-state interface shape remains close to sinusoidal as the higher harmonics are damped by the smoothing effect of surface tension. This is demonstrated by both the numerical simulations and an approximate calculation for small  $\delta$ .

Stability results are obtained using the Floquet analysis, which allows us to reduce the problem to the domain of the size of the spatial scale of the charge density variation. The effect of substrate patterning can be either stabilizing or destabilizing, depending on the parameter values. A stabilizing effect is observed at higher values of the pattern wavelength and has a nonmonotonic dependence on the amplitude of the substrate charge density variation. For small  $\delta$ , the correction to the stability criterion increases rapidly, consistent with the predictions made at small  $\delta$ . However, at larger values of  $\delta$ , typically above 0.2, the stability correction actually decreases, a counterintuitive result not reported in any of the previous studies of the stability of electrolyte films on patterned or structured surfaces.

## ACKNOWLEDGMENT

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