Appendix B

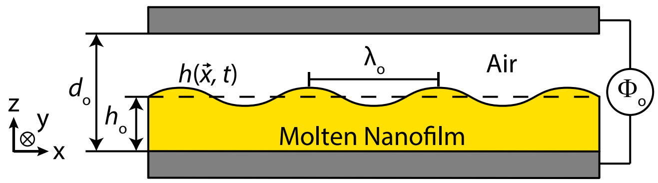
EVALUATION OF DRIVING FIELDS AT PERTURBED INTERFACES

B.1 Background

One interesting aspect of the derivation presented in Ch. 2 is that the driving mechanisms, the electric field for the SC model and the temperature field for the AP and TC models, are evaluated at the perturbed film interface $h(x, y, t)$, not the unperturbed film interface h_o. Intuitively, this is reasonable because the force due to surface tension is based on the perturbed surface since there is no curvature in the unperturbed state. To balance a driving force against surface tension, it should be calculated in the same configuration where the surface tension was calculated. One method for calculating the driving force is to use perturbation theory where the perturbed fields are typically evaluated at the base state, not the perturbed state. Evaluating some elements at the perturbed interface while evaluating others at the unperturbed interface leads to a subtle dissonance in the derivation which results in errors. This appendix will detail an example to show the error that can occur if the driving forces are not consistently evaluated at the perturbed interface.

The derivation of the SC model that was presented in Sec. 2.3.1 is actually a subset of the full derivation presented in the Ph.D. thesis of Zhuang [3]. In his original derivation, he considered a more general system in which an overall potential difference was applied across the nanofilm/air bilayer in addition to the surface charge present at the interface. This applied potential difference was not applicable to the experimental setup presented above, so the applied potential difference was not included in Ch. 2. However, the case where there is no interfacial charge density and only an applied potential difference has been investigated as a separate instability, called the electrohydrodynamic (EHD) instability. The EHD instability has garnered considerable interest both experimentally [4, 5, 75] and theoretically [76, 77]. As shown in Fig. B.1, the geometry is the same as in Ch. 2 except instead of an applied temperature gradient, there is an applied electric field. This appendix will focus on the theoretical work of Pease and Russel [76, 77] and demonstrate that their electric fields yield a tangential stress which is incompatible with Maxwell's equations.

Figure B.1: Instability geometry in EHD model



The driving force in the EHD model is the applied potential difference across the bilayer, Φ_o.

The rest of this appendix is organized as follows. In Sec. B.2, it is shown that there are no tangential stresses at a perfect dielectric interface with no free charge which arises directly from Maxwell's equation. Then, electric fields in the bilayer are derived from the nondimensional governing equations within the lubrication approximation when evaluated at the perturbed interface in Sec. B.3. Next, the tangential stresses due to these electric fields are computed in Sec. B.4 to show that the tangential stresses at the perturbed interface are zero. Then, a dimensional perturbation calculation follows to evaluate the perturbed electric field at the unperturbed interface in Sec. B.5. In Sec. B.6, it is demonstrated that these electric fields do not consistently satisfy Maxwell's equations and in Sec. B.7 the results are briefly discussed.

B.2 Tangential Stresses at a Perfect Dielectric Interface

The Maxwell stress tensor in the absence of magnetic fields, originally defined in Eq. (2.49), has the form

$$\mathbf{T}^{\text{em}} = \vec{E}\vec{D} - \frac{1}{2}\mathbf{I}\left(\vec{E}\cdot\vec{D}\right). \tag{B.1}$$

To compute the tangential stresses at the interface, this equation is dotted by the tangential unit vector, $\hat{t}$, on the left, dotted by the normal unit vector, $\hat{n}$, on the right, and the difference between the stress tensors in the air and film layers is taken. Note that all of these terms are evaluated at the film/air interface.

$$\hat{t}\cdot\mathbf{T}^{\text{em}}_{\text{air}}\cdot\hat{n} - \hat{t}\cdot\mathbf{T}^{\text{em}}_{\text{film}}\cdot\hat{n} = \hat{t}\cdot\vec{E}_{\text{air}}\vec{D}_{\text{air}}\cdot\hat{n} - \hat{t}\cdot\vec{E}_{\text{film}}\vec{D}_{\text{film}}\cdot\hat{n}.$$

The normal and tangential unit vectors are orthogonal so that $\hat{t}\cdot\mathbf{I}\cdot\hat{n} = 0$ which was used to simplify the preceding equation. The tangential components of the electric field must be equal across the interface because $\boldsymbol{\nabla}\times\vec{E} = 0$. Therefore, the subscripts on the electric field terms are dropped and the common terms factored out front

$$\hat{t}\cdot\mathbf{T}^{\text{em}}_{\text{air}}\cdot\hat{n} - \hat{t}\cdot\mathbf{T}^{\text{em}}_{\text{film}}\cdot\hat{n} = \left(\hat{t}\cdot\vec{E}\right)\left(\vec{D}_{\text{air}}\cdot\hat{n} - \vec{D}_{\text{film}}\cdot\hat{n}\right).$$

The difference in the normal components of the electric displacement field across the interface is simply the free charge at the interface, σ_{free}. In this system, there is no free charge, so the tangential stresses at the interface must be

$$\hat{t} \cdot \mathbf{T}^{\text{em}}_{\text{air}} \cdot \hat{n} - \hat{t} \cdot \mathbf{T}^{\text{em}}_{\text{film}} \cdot \hat{n} = \left(\hat{t} \cdot \vec{E}\right) \sigma_{\text{free}} = 0. \tag{B.2}$$

The fact that there can be no tangential stresses at a perfect dielectric interface if there is no free charge was well known to the leaky dielectric community [78]. As such, Pease and Russel did not calculate the tangential stresses at the perturbed interface in their derivation. However, as shown below in Sec. B.5 and Sec. B.6, their electric field expressions did cause tangential stresses anytime the interface was perturbed from the initially flat state. Before this is done, an electric field solution which is self-consistent with Maxwell's equations at the perturbed film interface will be demonstrated.

B.3 Electric Field Evaluated at a Perturbed Interface

The basics of the governing equations for the electric field within the lubrication approximation have been presented in Sec. 2.3.1, although in this section the potential scaling and the boundary conditions will be slightly different. All scaled quantities in this appendix will be denoted with a prime to signify the change in scaling. The characteristic potential scale is now

$$\Phi'_c = \Phi_o. \tag{B.3}$$

The electrostatic boundary conditions are changed to

$$\widetilde{\phi}'_{\text{film}}(Z = 0) = 0, \tag{B.4}$$

$$\widetilde{\phi}'_{\text{air}}(Z = D) = 1, \tag{B.5}$$

$$\widetilde{\phi}'_{\text{film}}(Z = H) = \widetilde{\phi}'_{\text{air}}(Z = H), \tag{B.6}$$

$$\varepsilon_{\text{film}} \frac{\partial \widetilde{\phi}'_{\text{film}}(Z = H)}{\partial Z} = \frac{\partial \widetilde{\phi}'_{\text{air}}(Z = H)}{\partial Z}. \tag{B.7}$$

The solutions for the electric potential have the same general solution

$$\widetilde{\phi}'_{\text{film}} = A^{\text{EHD}}_{\text{film}} Z + B^{\text{EHD}}_{\text{film}},$$
$$\widetilde{\phi}'_{\text{air}} = A^{\text{EHD}}_{\text{air}} Z + B^{\text{EHD}}_{\text{air}},$$

where $A_{\text{film}}^{\text{EHD}}$, $B_{\text{film}}^{\text{EHD}}$, $A_{\text{air}}^{\text{EHD}}$, and $B_{\text{air}}^{\text{EHD}}$ are integration constants. Applying the Dirichlet boundary conditions from Eq. (B.4) and (B.5) yields

$$\widetilde{\phi}'_{\text{film}} = A_{\text{film}}^{\text{EHD}} Z,$$
$$\widetilde{\phi}'_{\text{air}} = A_{\text{air}}^{\text{EHD}}(Z - D) + 1.$$

Applying Eq. (B.7) gives

$$\varepsilon_{\text{film}} A_{\text{film}}^{\text{EHD}} = A_{\text{air}}^{\text{EHD}}.$$

This implies that the electric potentials become

$$\widetilde{\phi}'_{\text{film}} = A_{\text{film}}^{\text{EHD}} Z,$$
$$\widetilde{\phi}'_{\text{air}} = \varepsilon_{\text{film}} A_{\text{film}}^{\text{EHD}}(Z - D) + 1.$$

The final boundary condition is continuity of the potentials at the interface from Eq. (B.6). This allows $A_{\text{film}}^{\text{EHD}}$ to be determined

$$A_{\text{film}}^{\text{EHD}} = \frac{1}{\varepsilon_{\text{film}} D - (\varepsilon_{\text{film}} - 1)H}.$$

The electric potentials are therefore

$$\widetilde{\phi}'_{\text{film}} = \frac{Z}{\varepsilon_{\text{film}} D - (\varepsilon_{\text{film}} - 1)H}, \tag{B.8}$$
$$\widetilde{\phi}'_{\text{air}} = \frac{\varepsilon_{\text{film}}(Z - D)}{\varepsilon_{\text{film}} D - (\varepsilon_{\text{film}} - 1)H} + 1. \tag{B.9}$$

From this the electric fields are computed and then broken into components

$$\widetilde{E}'_{\text{film,z}} = \frac{-1}{\varepsilon_{\text{film}} D - (\varepsilon_{\text{film}} - 1)H}, \tag{B.10}$$
$$\widetilde{E}'_{\text{film},\|} = \frac{-Z(\varepsilon_{\text{film}} - 1)\epsilon \widetilde{\boldsymbol{\nabla}}_{\|} H}{[\varepsilon_{\text{film}} D - (\varepsilon_{\text{film}} - 1)H]^2}, \tag{B.11}$$
$$\widetilde{E}'_{\text{air,z}} = \frac{-\varepsilon_{\text{film}}}{\varepsilon_{\text{film}} D - (\varepsilon_{\text{film}} - 1)H}, \tag{B.12}$$
$$\widetilde{E}'_{\text{air},\|} = \frac{-\varepsilon_{\text{film}}(Z - D)(\varepsilon_{\text{film}} - 1)\epsilon \widetilde{\boldsymbol{\nabla}}_{\|} H}{[\varepsilon_{\text{film}} D - (\varepsilon_{\text{film}} - 1)H]^2}. \tag{B.13}$$

B.4 Tangential Stresses from Electric Field Evaluated at Perturbed Interface

In Sec. 2.3.1, an expression for the normal component of the stress tensor dotted into the normal vector was computed. Recall that this expression is

$$\hat{n} \cdot \mathbf{T}^{\text{em}} \cdot \hat{n} = \frac{\varepsilon_o \varepsilon}{2} E_z^2. \tag{B.14}$$

To find the tangential stresses this quantity must be subtracted from the stress tensor dotted into the normal vector. This quantity is

$$\mathbf{T}^{\mathrm{em}}\cdot\hat{n} = \varepsilon_o\varepsilon \begin{bmatrix} -\frac{1}{2}E_z^2 & 0 & E_xE_z \\ 0 & -\frac{1}{2}E_z^2 & E_yE_z \\ E_xE_z & E_yE_z & \frac{1}{2}E_z^2 \end{bmatrix} \begin{bmatrix} -\epsilon\frac{\partial H}{\partial X} \\ -\epsilon\frac{\partial H}{\partial Y} \\ 1 \end{bmatrix} = \varepsilon_o\varepsilon\left(\frac{\epsilon}{2}E_z^2\widetilde{\boldsymbol{\nabla}}_{\|}H + E_z\vec{E}_{\|} + \frac{1}{2}E_z^2\hat{z}\right).$$

The tangential components of the stress tensor are

$$\begin{aligned} \left(\mathbf{T}^{\mathrm{em}}\cdot\hat{n}\right)_{\|} &= \mathbf{T}^{\mathrm{em}}\cdot\hat{n} - \left(\hat{n}\cdot\mathbf{T}^{\mathrm{em}}\cdot\hat{n}\right)\hat{n} \\ &= \varepsilon_o\varepsilon E_z\left(E_z\epsilon\widetilde{\boldsymbol{\nabla}}_{\|}H + \vec{E}_{\|}\right). \end{aligned} \tag{B.15}$$

The difference in the tangential stress tensors in the air and the film is now verified to be zero. This implies that there are no tangential stresses.

$$\begin{aligned} \left(\mathbf{T}^{\mathrm{em}}_{\mathrm{air}}\cdot\hat{n}\right)_{\|} - \left(\mathbf{T}^{\mathrm{em}}_{\mathrm{film}}\cdot\hat{n}\right)_{\|} =& \varepsilon_o E'_{\mathrm{air,z}}\left(E'_{\mathrm{air,z}}\epsilon\widetilde{\boldsymbol{\nabla}}_{\|}H + \vec{E}'_{\mathrm{air},\|}\right) \\ & - \varepsilon_o\varepsilon_{\mathrm{film}}E'_{\mathrm{film,z}}\left(\epsilon E'_{\mathrm{film,z}}\widetilde{\boldsymbol{\nabla}}_{\|}H + \vec{E}'_{\mathrm{film},\|}\right) \\ =& \frac{\varepsilon_o\varepsilon_{\mathrm{film}}\Phi_c'^2}{h_o^2}\widetilde{E}'_{\mathrm{film,z}}\left(\epsilon\widetilde{\boldsymbol{\nabla}}_{\|}H\left(\widetilde{E}'_{\mathrm{air,z}} - \widetilde{E}'_{\mathrm{film,z}}\right) + \widetilde{E}'_{\mathrm{air},\|} - \widetilde{E}'_{\mathrm{film},\|}\right). \end{aligned}$$

In this expression the fact that $E'_{\mathrm{air,z}} = \varepsilon_{\mathrm{film}}E'_{\mathrm{film,z}}$ has been used. Substitution of the electric field expressions into this equation yields

$$\begin{aligned} \left(\mathbf{T}^{\mathrm{em}}_{\mathrm{air}}\cdot\hat{n}\right)_{\|} - \left(\mathbf{T}^{\mathrm{em}}_{\mathrm{film}}\cdot\hat{n}\right)_{\|} =& \frac{\varepsilon_o\varepsilon_{\mathrm{film}}\Phi_c'^2}{h_o^2}\widetilde{E}'_{\mathrm{film,z}}\epsilon\widetilde{\boldsymbol{\nabla}}_{\|}H\left(\frac{1-\varepsilon_{\mathrm{film}}}{\varepsilon_{\mathrm{film}}D - (\varepsilon_{\mathrm{film}}-1)H}\right. \\ & \left. + \frac{H(\varepsilon_{\mathrm{film}}-1) - \varepsilon_{\mathrm{film}}(H-D)(\varepsilon_{\mathrm{film}}-1)}{\left[\varepsilon_{\mathrm{film}}D - (\varepsilon_{\mathrm{film}}-1)H\right]^2}\right) \\ =& 0. \end{aligned}$$

This demonstrates that the electric fields derived from the perturbed interface consistently satisfy Maxwell's equations and do not have any tangential stresses when there is no free charge at the interface.

B.5 Electric Field Perturbations Evaluated at an Unperturbed Interface

The derivation of Pease and Pussel computed the electric fields in the bilayer in two steps. First, a base state electric field was computed for the unperturbed film interface, within the geometry shown in Fig. B.1. Then, a perturbation to the film height was introduced which created perturbations in the electric fields. Their derivation proceeded in dimensional quantities and was then scaled after computation of the perturbed electric field.

B.5.1 Base State Electric Field

Base state quantities are denoted with the superscript ^{o}. The boundary conditions for this system are

$$\phi^o_{\text{film}}(z=0) = 0, \tag{B.16}$$

$$\phi^o_{\text{air}}(z=d_o) = \Phi_o, \tag{B.17}$$

$$\varepsilon_{\text{film}} E^o_{\text{film}}(z=h_o) = E^o_{\text{air}}(z=h_o), \tag{B.18}$$

$$\phi^o_{\text{air}}(z=d_o) - \phi^o_{\text{film}}(z=0) = -\int_0^{h_o} E^o_{\text{film}} dz - \int_{h_o}^{d_o} E^o_{\text{air}} dz. \tag{B.19}$$

The last condition is an equivalent statement to the continuity of electric potential at an interface which arises from the tangential electrostatic boundary conditions. Combining the four equations presented above into one simplifies to

$$\Phi_o = -h_o E^o_{\text{film}} - \varepsilon_{\text{film}}(d_o - h_o) E^o_{\text{film}},$$

from which the electric field in the film at $z = h_o$ was found

$$\vec{E}^o_{\text{film}}(z=h_o) = \frac{-\Phi_o \hat{z}}{\varepsilon_{\text{film}} d_o - (\varepsilon_{\text{film}} - 1) h_o}. \tag{B.20}$$

From this expression the electric field in the air layer at $z = h_o$ was computed to be

$$\vec{E}^o_{\text{air}}(z=h_o) = \frac{-\varepsilon_{\text{film}} \Phi_o \hat{z}}{\varepsilon_{\text{film}} d_o - (\varepsilon_{\text{film}} - 1) h_o}. \tag{B.21}$$

These are the base state electric fields which can now be perturbed.

B.5.2 Perturbed Electric Field

With the base state electric fields computed, the position of the film/air interface was perturbed

$$h = h_o + \delta h e^{i\vec{k}_\parallel \cdot \vec{x}_\parallel}. \tag{B.22}$$

The perturbed electric quantities, $\delta\phi$, $\delta\vec{E}$, and $\delta\vec{D}$ are added to the their respective base state quantities. For each layer, the total potential, electric field, and electric displacement field must satisfy Maxwell's equations. Due to the linearity of Maxwell's equations the perturbed electric fields must satisfy

$$\nabla \times \delta\vec{E} = 0, \tag{B.23}$$

$$\nabla \cdot \delta\vec{E} = 0. \tag{B.24}$$

These two equations imply that the perturbed electric field can be written as the negative gradient of the perturbed potential and that this perturbed potential will satisfy Laplace's equation

$$\nabla^2 \delta\phi = 0. \tag{B.25}$$

The perturbed potential was expanded in terms of normal modes as was the film height perturbation. The specific form is

$$\delta\phi = \widetilde{\phi}(z) e^{i\vec{k}_{\parallel}\cdot\vec{x}_{\parallel}}. \tag{B.26}$$

Laplace's equation of the perturbed potential then becomes

$$\frac{d^2\widetilde{\phi}}{dz^2} - k^2\widetilde{\phi} = 0. \tag{B.27}$$

Two linearly independent solutions to this equation in the two layers are

$$\widetilde{\phi}_{\text{film}} = A^{\text{EHD}}_{\text{film}} \sinh kz + B^{\text{EHD}}_{\text{film}} \cosh kz, \tag{B.28}$$

$$\widetilde{\phi}_{\text{air}} = A^{\text{EHD}}_{\text{air}} \sinh kz + B^{\text{EHD}}_{\text{air}} \cosh kz. \tag{B.29}$$

Because the base state solution already satisfies the boundary conditions at $z = 0$ and $z = d_o$, the perturbed potential must satisfy the following Dirichlet boundary conditions

$$\delta\phi_{\text{film}}(z = 0) = 0, \tag{B.30}$$

$$\delta\phi_{\text{air}}(z = d_o) = 0. \tag{B.31}$$

Upon simplification the perturbed potentials become

$$\widetilde{\phi}_{\text{film}} = A^{\text{EHD}}_{\text{film}} \sinh kz,$$

$$\widetilde{\phi}_{\text{air}} = A^{\text{EHD}}_{\text{air}} \sinh k(z - d_o),$$

where in the last expression a constant factor has been absorbed into the definition of $A_{\text{air}}^{\text{EHD}}$ since it would have canceled later. The two remaining boundary conditions are more complicated to apply and in the work of Pease and Russel were evaluated at the unperturbed film position $z = h_o$, even though it should have been $z = h$. Denoting the difference across the interface (air minus film) of a quantity by enclosing it in brackets, the usual electrostatic boundary conditions are

$$\hat{n} \times [\vec{E}] = 0, \tag{B.32}$$

$$\hat{n} \cdot [\vec{D}] = 0. \tag{B.33}$$

The vector quantities are now broken into components to more effectively take the dot and cross product in the above equations. The base state electric field has no $\hat{x}$ and $\hat{y}$ components, and the normal component of the base state electric displacement field is continuous across the interface because there is no free charge. Consequently the differences across the interface have the form

$$\begin{aligned}
[E_x] &= [\delta E_x], \\
[E_y] &= [\delta E_y], \\
[E_z] &= [E^o] + [\delta E_z], \\
[D_x] &= [\delta D_x], \\
[D_y] &= [\delta D_y], \\
[D_z] &= [\delta D_z].
\end{aligned}$$

The boundary condition in the normal direction is

$$\hat{n} \cdot [\vec{D}] = -\frac{\partial \delta h}{\partial x}[\delta D_x] - \frac{\partial \delta h}{\partial y}[\delta D_y] + [\delta D_z] = 0.$$

The first two terms in this expression are second order and have been dropped to first order. Consequently, this equation implies

$$\delta E_{\text{air},z} = \varepsilon_{\text{film}} \delta E_{\text{film},z}.$$

Or, in terms of the potential,

$$\frac{\partial \widetilde{\phi}_{\text{air}}}{\partial z} = \varepsilon_{\text{film}} \frac{\partial \widetilde{\phi}_{\text{film}}}{\partial z}.$$

This equation was evaluated at $z = h_o$ and yields one constant in terms of the other

$$A_{\text{film}}^{\text{EHD}} = \frac{A_{\text{air}}^{\text{EHD}}}{\varepsilon_{\text{film}}} \frac{\cosh k(h_o - d_o)}{\cosh kh_o}, \tag{B.34}$$

which means that the perturbed potentials are now

$$\begin{aligned} \widetilde{\phi}_{\text{film}} &= \frac{A_{\text{air}}^{\text{EHD}} \cosh k(h_o - d_o)}{\varepsilon_{\text{film}}} \frac{\sinh kz}{\cosh kh_o}, \\ \widetilde{\phi}_{\text{air}} &= A_{\text{air}}^{\text{EHD}} \sinh k(z - d_o). \end{aligned}$$

The tangential electrostatic boundary conditions requires the evaluation of the cross product

$$\hat{n} \times [\vec{E}] = \begin{bmatrix} \hat{x} & \hat{y} & \hat{z} \\ -\frac{\partial h}{\partial x} & -\frac{\partial h}{\partial y} & -1 \\ [\delta E_x] & [\delta E_y] & [E^o] + [\delta E_z] \end{bmatrix} = 0.$$

The $\hat{z}$ component of this cross product is second order and the $\hat{x}$ and $\hat{y}$ expressions have the same form. As such, the $\hat{x}$ component was chosen without loss of generality. In terms of the potential it is

$$\left[-\frac{\partial \delta\phi}{\partial y} + E^o \frac{\partial h}{\partial y} \right] = 0.$$

Because both the potential perturbation and the height perturbation were expanded in the same set of normal modes, the partial derivative brings down the same term from the exponential which then cancels, leaving

$$\left[-\widetilde{\phi} + E^o \delta h \right] = 0.$$

The difference in perturbed potentials is then

$$\widetilde{\phi}_{\text{film}} - \widetilde{\phi}_{\text{air}} = \delta h \left(E_{\text{film}}^o - E_{\text{air}}^o \right). \tag{B.35}$$

From this equation the last remaining constant in the perturbed potentials is determined

$$A_{\text{air}}^{\text{EHD}} = \frac{\varepsilon_{\text{film}}}{\cosh k(h_o - d_o)} \frac{\delta h \left(E_{\text{film}}^o - E_{\text{air}}^o \right)}{\tanh kh_o - \varepsilon_{\text{film}} \tanh k(h_o - d_o)}. \tag{B.36}$$

Substituting this back into the expressions for the perturbed potentials yields

$$\widetilde{\phi}_{\text{film}} = \frac{\delta h(\varepsilon_{\text{film}} - 1)E^o_{\text{film}}}{\varepsilon_{\text{film}} \tanh k(h_o - d_o) - \tanh kh_o} \frac{\sinh kz}{\cosh kh_o}, \tag{B.37}$$

$$\widetilde{\phi}_{\text{air}} = \frac{\varepsilon_{\text{film}}\delta h(\varepsilon_{\text{film}} - 1)E^o_{\text{film}}}{\varepsilon_{\text{film}} \tanh k(h_o - d_o) - \tanh kh_o} \frac{\sinh k(z - d_o)}{\cosh k(h_o - d_o)}. \tag{B.38}$$

The components of the electric field at the perturbed interface ($z = h$) are needed for the evaluation of the tangential stresses. As before, the electric field is broken into components normal and tangential to the interface

$$\delta E_{\text{film,z}} = \frac{-\delta h(\varepsilon_{\text{film}} - 1)E^o_{\text{film}}}{\varepsilon_{\text{film}} \tanh k(h_o - d_o) - \tanh kh_o} \frac{k \cosh kh}{\cosh kh_o}, \tag{B.39}$$

$$\delta E_{\text{film},\parallel} = \frac{-(\varepsilon_{\text{film}} - 1)E^o_{\text{film}}\nabla_\parallel \delta h}{\varepsilon_{\text{film}} \tanh k(h_o - d_o) - \tanh kh_o} \frac{\sinh kh}{\cosh kh_o}, \tag{B.40}$$

$$\delta E_{\text{air,z}} = \frac{\varepsilon_{\text{film}}\delta h(\varepsilon_{\text{film}} - 1)E^o_{\text{film}}}{\varepsilon_{\text{film}} \tanh k(h_o - d_o) - \tanh kh_o} \frac{k \cosh k(h - d_o)}{\cosh k(h_o - d_o)}, \tag{B.41}$$

$$\delta E_{\text{air},\parallel} = \frac{-\varepsilon_{\text{film}}(\varepsilon_{\text{film}} - 1)E^o_{\text{film}}\nabla_\parallel \delta h}{\varepsilon_{\text{film}} \tanh k(h_o - d_o) - \tanh kh_o} \frac{\sinh k(h - d_o)}{\cosh k(h_o - d_o)}. \tag{B.42}$$

Since these electric fields are in dimensional units, they are nondimensionalized and then the lubrication approximation is applied to them in the following equations. During the application of the lubrication approximation the hyperbolic functions are Taylor expanded. Note that all dependence on the wavevector, K, cancels after the Taylor expansion.

$$\widetilde{\delta E}'_{\text{film,z}} = \frac{-(H-1)(\varepsilon_{\text{film}}-1)\widetilde{E}^o_{\text{film}}}{\varepsilon_{\text{film}}\tanh\epsilon K(1-D)-\tanh\epsilon K}\frac{\epsilon K\cosh\epsilon KH}{\cosh\epsilon K}$$
$$\approx (H-1)\frac{(\varepsilon_{\text{film}}-1)\widetilde{E}^o_{\text{film}}}{\varepsilon_{\text{film}}D-(\varepsilon_{\text{film}}-1)}, \tag{B.43}$$
$$\widetilde{\delta E}'_{\text{film},\parallel} = \frac{-(\varepsilon_{\text{film}}-1)\widetilde{E}^o_{\text{film}}\epsilon\widetilde{\boldsymbol{\nabla}}_\parallel H}{\varepsilon_{\text{film}}\tanh\epsilon K(1-D)-\tanh\epsilon K}\frac{\sinh\epsilon KH}{\cosh\epsilon K}$$
$$\approx \epsilon H\widetilde{\boldsymbol{\nabla}}_\parallel H\frac{(\varepsilon_{\text{film}}-1)\widetilde{E}^o_{\text{film}}}{\varepsilon_{\text{film}}D-(\varepsilon_{\text{film}}-1)}, \tag{B.44}$$
$$\widetilde{\delta E}'_{\text{air,z}} = \frac{-(H-1)\varepsilon_{\text{film}}(\varepsilon_{\text{film}}-1)\widetilde{E}^o_{\text{film}}}{\varepsilon_{\text{film}}\tanh\epsilon K(1-D)-\tanh\epsilon K}\frac{\epsilon K\cosh\epsilon KH}{\cosh\epsilon K}$$
$$\approx (H-1)\frac{\varepsilon_{\text{film}}(\varepsilon_{\text{film}}-1)\widetilde{E}^o_{\text{film}}}{\varepsilon_{\text{film}}D-(\varepsilon_{\text{film}}-1)}, \tag{B.45}$$
$$\widetilde{\delta E}'_{\text{air},\parallel} = \frac{-\varepsilon_{\text{film}}(\varepsilon_{\text{film}}-1)\widetilde{E}^o_{\text{film}}\epsilon\widetilde{\boldsymbol{\nabla}}_\parallel H}{\varepsilon_{\text{film}}\tanh\epsilon K(1-D)-\tanh\epsilon K}\frac{\sinh\epsilon K(H-D)}{\cosh\epsilon K}$$
$$\approx \epsilon(H-D)\widetilde{\boldsymbol{\nabla}}_\parallel H\frac{\varepsilon_{\text{film}}(\varepsilon_{\text{film}}-1)\widetilde{E}^o_{\text{film}}}{\varepsilon_{\text{film}}D-(\varepsilon_{\text{film}}-1)}. \tag{B.46}$$

Additionally, the scaled base state electric fields at the unperturbed interface are

$$\widetilde{E}^o_{\text{film}} = \frac{-1}{\varepsilon_{\text{film}}D-(\varepsilon_{\text{film}}-1)}, \tag{B.47}$$
$$\widetilde{E}^o_{\text{air}} = \frac{-\varepsilon_{\text{film}}}{\varepsilon_{\text{film}}D-(\varepsilon_{\text{film}}-1)}. \tag{B.48}$$

B.6 Tangential Stresses from Electric Field Evaluated at Unperturbed Interface

From Sec. B.4 the tangential components of the stress tensor have the form

$$(\mathbf{T}^{\text{em}}\cdot\hat{n})_\parallel = \varepsilon_o\varepsilon\left(\epsilon E_z^2\widetilde{\boldsymbol{\nabla}}_\parallel H + \vec{E}_\parallel E_z\right). \tag{B.49}$$

The $\hat{z}$ component of the electric field is composed of a base state and a perturbation. Substituting and only keeping terms to first order in ϵ, this expression becomes

$$(\mathbf{T}^{\text{em}}\cdot\hat{n})_\parallel = \varepsilon_o\varepsilon\left(\epsilon\widetilde{\boldsymbol{\nabla}}_\parallel H(E^o)^2 + \vec{E}_\parallel E^o\right).$$

This implies that the tangential stress difference between the air and film layers is

$$\left(\mathbf{T}^{\text{em}}_{\text{air}}\cdot\hat{n}\right)_\parallel - \left(\mathbf{T}^{\text{em}}_{\text{film}}\cdot\hat{n}\right)_\parallel = \frac{\varepsilon_o\Phi_c'^2}{h_o^2}\varepsilon_{\text{film}}\widetilde{E}^o_{\text{film}}\Big(\epsilon\widetilde{\boldsymbol{\nabla}}_\parallel H\widetilde{E}^o_{\text{film}}(\varepsilon_{\text{film}}-1)$$
$$+\widetilde{\delta E}'_{\text{air},\parallel} - \widetilde{\delta E}'_{\text{film},\parallel}\Big).$$

Focusing on the right hand side of the equation, substitution of the scaled electric fields from above gives

$$\begin{aligned}\left(\mathbf{T}^{\text{em}}_{\text{air}} \cdot \hat{n}\right)_{\parallel} - \left(\mathbf{T}^{\text{em}}_{\text{film}} \cdot \hat{n}\right)_{\parallel} =& \frac{\varepsilon_o \varepsilon_{\text{film}} \Phi_c'^2}{h_o^2} \left(\widetilde{E}^o_{\text{film}}\right)^2 \epsilon \widetilde{\boldsymbol{\nabla}_{\parallel}} H(\varepsilon_{\text{film}} - 1) \times \\ & \left(1 - \frac{\varepsilon_{\text{film}} D - (\varepsilon_{\text{film}} - 1) H}{\varepsilon_{\text{film}} D - (\varepsilon_{\text{film}} - 1)}\right) \\ \neq& 0. \end{aligned} \tag{B.50}$$

This result demonstrates that this technique for calculating the electric field is not consistent with Maxwell's equations any time that the interface is not flat ($H = 1$).

B.7 Summary

In the first case when the electric field was evaluated at the perturbed interface, the governing equations were scaled and the lubrication approximation was invoked very early in the derivation. This use of the lubrication approximation simplified Laplace's equation, which allowed for an easy solution of the electric field in the bilayer since the $\hat{z}$ equations decouple from the other directions. In the second case, the electric field was computed in dimensional quantities, scaled, and then the lubrication approximation was applied afterwards. This leads to a more complicated solution process which can obscure the fact the electric fields are not consistent with Maxwell's equations any time the interface is not flat. From an intuitive perspective, the choice to evaluate the perturbed electric field at the unperturbed interface is problematic because it attempts to find a consistent electric field at a flat interface when the interface will be deformed during growth. The computation of the electric fields in the perturbed case could have been fixed by applying the final two electrostatic boundary conditions at $z = h$ instead of $z = h_o$.

As an interesting historical note, this issue with the inconsistent tangential stresses was corrected in later work (e.g. [79]) without comment, so it is not clear if this issue was ever noticed. It also bears mentioning that even with this error Pease and Russel still derived the correct expression for λ_o^{EHD} in linear stability (not presented here). They found the correct answer because in linear stability the stresses are evaluated at the unperturbed interface where the tangential stresses don't contribute. This means that the only place where this issue would cause significant problems is in numerical simulations of the EHD thin film evolution equation at late times when using the evolution equation derived by Pease and Russel.