Nonlinear interfacial stability of core-annular film flows in the presence of surfactants

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ABSTRACT

NONLINEAR INTERFACIAL STABILITY OF CORE-ANNULAR FILM FLOWS IN THE PRESENCE OF SURFACTANTS

by

Said A. Kas-Danouche

This work is an analytical and computational study of the nonlinear interfacial instabilities found in core-annular flows in the presence of surfactants. Core-annular flows arise when two immiscible fluids (for example water and oil) are caused to flow in a pipe under the action of an axial pressure gradient. In one typical type of flow regime, the fluids arrange themselves so that the less viscous (e.g. water) lies in the region of high shear near the pipe wall, with the more viscous fluid occupying the core region. Technologically, this arrangement provides an advantage since the highly viscous fluid is lubricated by the less viscous annulus and for a given pressure gradient the core-fluid flux can be greatly increased.

The stability of these flows is of fundamental scientific and practical importance. The sharp interface between the two phases can become unstable by several physical mechanisms and one such mechanism of practical importance is surface tension. In this work we incorporate into our model the effects of insoluble surfactants on the instability. The full problem is derived with particular emphasis paid to the surfactant transport equation which is novel. We then carry out an asymptotic solution of the problem when the annular layer is thin compared to the core-fluid radius and for waves which are of the order of the pipe radius (that is long compared to the annular layer thickness); these scales are in accord with both linear theory as well as experimental observations. The result of the matched asymptotic analysis is a system of coupled nonlinear partial differential equations for the interfacial amplitude and the surfactant concentration on the interface. In the absence of surfactants, the system reduces to the Kuramoto-Sivashinsky equation which has been extensively studied.
as a paradigm for one-dimensional turbulence in dissipative systems. The surfactant modifies the flow by inducing Marangoni forces along the interface which in turn modify both the velocities and interfacial amplitudes. There are two parameters present in the nonlinear system, the length of the system and a surface Peclet number which measures the diffusion of surfactant on the interface.

In order to gain an understanding of the dynamics, we carry out extensive computations using accurate and stable numerical methods capable of following the solution for long times. We map out the dynamics by numerically solving initial value problems on spatially periodic domains where the length of the system is the bifurcation parameter, keeping the Peclet number fixed and equal to one. We find that surfactant acts to suppress chaotic behavior found in its absence for extensive ranges of the bifurcation parameter. The new flow consists of successive windows (in parameter space) of steady-state traveling waves separated by time-periodic attractors. As the length of the system increases a self-similar structure has been found to govern the shapes of the traveling waves as we move from a given window to a lower one. This is elucidated analytically and numerically.
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FILM FLOWS IN THE PRESENCE OF SURFACTANTS

by
Said A. Kas-Danouche

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S. A. Kas-Danouche, D. T. Papageorgiou, and M. Siegel
“Nonlinear interfacial stability of core-annular film flows in the presence of surfactants”,
To the Almighty God, the God of Abraham, Isaac, and Jacob
To my beloved wife, Patricia
To my wonderful mom, Yermy
To the memory of my dad, Georges
To my sons, Jorge, Said, and Yamil

This is my beloved family

A el Dios Todopoderoso, el Dios de Abraham, Isaac y Jacob
A mi amada esposa, Patricia
A mi maravillosa mamá, Yermy
A la memoria de mi papá, Georges
A mis hijos, Jorge, Said y Yamil
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A.1 Radius ($R_1$) of the circumference circumscribed to the interface in a plane parallel to the unit normal vector, $\vec{n}$, and forming an angle $\theta$ with the cylinder axis.

A.2 Radius ($R_2$) of the circumference circumscribed to the interface and parallel to the cylinder axis. $\vec{n}$ is the unit normal vector and $\vec{t}$ is the tangential unit vector with slope $S'$, of the interface; and $\theta$ is the angle formed between $\vec{n}$ and the cylinder axis.
Two immiscible fluids, generally, arrange themselves such that the less viscous one is in the region of high shear (Joseph and Renardy [38], 1993). So, it could be possible to introduce a lubricating fluid with a beneficial effect in a flow of a very viscous fluid. This suggests that lubricated flows are stable. Also, when a fluid in a capillary cylinder is displaced by another, a layer of the first fluid is left behind coating the cylinder walls (Taylor [74], 1961). This is called a core-annular flow. In general, core-annular flows are parallel flows of immiscible liquids in a cylinder; one fluid flows through the cylinder core and the other ones move in successive annuli that surround the core fluid. Core-annular flow occurs, for example, during liquid-liquid displacements in porous media (Edwards, Brenner, and Wasan [16], 1991) when a wetting layer is present, and in lung airways where internal airway surface is coated with a thin liquid lining (Halpern and Grotberg [24], 1992 and [25], 1993).

**Figure 1.1** Cartoons of core-annular flows of water-lubricated oil when the oil is lighter than water. (a) is oil in water, (b) bubbles of oil in water, (c), (d) slugs of oil, (e) stratified flow, (f) corkscrew core flow, (g), (h) flying core flow, and (i) water in oil. Taken from D. D. Joseph, R. Bai, K. P. Chen, and Y. Y. Renardy. ([34], 1997)
The case of lubricated pipelining is an important technological application of interest to the oil industry, where the annular liquid (water) lubricates the motion of the core liquid (viscous oil). There are several flow regimes in horizontal pipes including, stratified flow with heavy fluid below, water in oil with(out) emulsions, bamboo waves, oil bubbles and slugs in water, and an annulus of water surrounding a concentric oil core. Figure 1.1 shows cartoons for some types of core-annular flows when the oil is lighter than water.

1.1 Experimental Results

In the case of two co-flowing fluids with a core-annular configuration, the first mention of water lubricated oil in pipelines emerges in a patent by Isaacs and Speed ([31], 1904). They discussed water lubrication of oil where the oil density is lower than the lubricating liquid density. Emulsification happens when the oil viscosity is less than 500 cp. Lubrication vanishes when water emulsifies into the oil, leading to the undesirable situation of the emulsion viscosity being greater than the viscosity of the oil alone (Joseph, Bai, Chen, and Renardy [34], 1997). One of the first works that addresses the problem of core-annular flows, with the oil density greater than the lubricating fluid, is that of Clark and Shapiro in ([10], 1949). Lubricated pipelining is acceptable for water-lubricated heavy oils whose viscosity is bigger than 500 cp and the density is approximately that of water, $\rho_{oil} > 0.9g/cm^3$. Leach ([45], 1957) considered several options for pipelines to transport 48,000 barrels per day of heavy oil from Temblador to Caripe, Venezuela, a distance of 148 kilometers. He rejected the water-lubricated pipelining because of the uncertainty concerning the formation of emulsions among other reasons.

The Shell Development Company, in California, addressed the problem of emulsification. Clifton and Handley in ([11], 1958) wrote “Method and Apparatus for Lubricating Pipe Lines”. When heavy oil and water pass through a pump, the
water is emulsified into the oil producing an emulsion of water and oil which is even more viscous than the oil alone. An objective of their invention was to essentially remove all the water from the core-annular flow before it enters the pump, in order to prevent emulsification. Glass ([22], 1961) of Esso Research and Engineering experimented with flowing oil and water through a 4 ft length and 1 cm inner diameter glass tubing. If the crude oil is sufficiently viscous, then the initial core (oil) will be easily sustained. For less viscous oil, the core breaks up, and as the oil viscosity is decreased, the core forms more and more blobs of oil. Stein ([71], 1978), and Oliemans, Ooms, Wu and Duývestin ([51], 1985) have developed experiments on water-lubricated pipelining. Advances of economically acceptable pipelines has been developed by The Shell Oil Company. About ten years ago, Maraven of PdVSA (Petróleos de Venezuela Sociedad Anónima) implemented a 60 kilometer pipeline for the transportation of water-lubricated heavy oil. Figure 1.2 shows a pipe with a core-annular flow, which is used to transport water-lubricated oil, at San Tome, Venezuela (Daniel D. Joseph and Yuriko Y. Renardy [38], 1993).

Figure 1.2 Core-annular flow of water-lubricated oil in a pipe of diameter 8 in. and test loop of length 1km at San Tome, Venezuela. Taken from Daniel D. Joseph and Yuriko Y. Renardy ([38], 1993), Plate V.2.3. This photograph was supplied by INTEVEP, S.A., an affiliate of Petróleos de Venezuela.
Aul and Olbricht ([4], 1990) developed experiments on liquid-liquid displacements of low-Reynolds-number, pressure-driven core-annular flow in a straight capillary tube. The annular film is thinner than the core, and the film viscosity much larger than the core viscosity. They conclude that the film is unstable for all conditions considered in the experiments. Experimental studies of lubricated pipelining of oil with water have also been developed by Bai, Chen, and Joseph ([5], 1992). In their studies they found bamboo waves for up-flow, disturbed bamboo waves for both up-flow and down-flow, and corkscrew waves in down-flow (Figure 1.1(f) shows a cartoon of a horizontal corkscrew wave).

Scoffoni, Lajeunesse, and Homsy ([65], 2001) performed experiments of the downward displacements of one miscible fluid by another lighter and less viscous one, in a vertical pipe. They considered the velocities to be sufficiently high to neglect diffusive effects and speculate that the instabilities observed are driven by the viscosity stratification and are similar to what occurs in core-annular flows of immiscible fluids.

1.2 Theoretical Results for Clean Interfaces

There are a number of studies concerned with core-annular flows when the interfaces are free of surfactant. We begin our literature review in §1.2.1 with the studies of problems with no flow, in §1.2.2 consider studies of core-annular flows, and finally in §1.2.3 we conclude with simulations concerned with clean interfaces.

1.2.1 No Flow Core-Annular Arrangements

In the case of no flow, studies of a long cylindrical thread of a viscous liquid suspended in a different unbounded fluid were developed by Tomotika ([76], 1935). Goren ([23], 1962) studied the linear stability of an annular film coating a wire or the inner surface of a cylinder, when the ambient or core fluid is inviscid. He considered an annular
film initially of uniform thickness and at rest. His results indicate that the film is unstable to infinitesimal sinusoidal disturbances. He showed that the fastest growing disturbance has a finite wavelength that is long compared with the film thickness.

Hammond ([26], 1983) extended Goren's study into the nonlinear regime. He developed a nonlinear analysis based on lubrication theory for the adjustment of a thin annular film under surface tension. He suggests that an initial sinusoidal disturbance of the interface may lead to the breakup of the film in the form of axisymmetric droplets or 'lenses' of the annular liquid separated by the core fluid. Later, Gauglitz and Radke ([20], 1988) developed an alternative approximation based on the Hammond analysis by including the exact curvature expression in the theory. In this way, the approximate equation reduces to the Young-Laplace equation (Adamson [2], 1976) for capillary statics in regions where the fluid is nearly static, and acts like the usual evolution equation when the film is thin. They found a critical thickness which demarks the transition between films that evolve into stable collars and those that break up to form liquid lenses.

1.2.2 Core-Annular Flows

Some analyses have been done for the more general case of two co-flowing fluids with a core-annular configuration. In what follows, we give a brief review of linear and nonlinear stability studies.

**Linear stability studies:** The linear theory of stability has been studied by several people. Hickox ([28], 1971) studied core-annular flow in a vertical pipe and used a long-wave approximation to show that the waves are always unstable, for both axisymmetric and asymmetric disturbances, when the more viscous fluid is in the annulus. Joseph, Renardy, and Renardy ([35], 1983 and [36], 1984) considered the problem of lubricated pipelining with the core viscosity greater than the annular film
viscosity and showed that the core-annular flows could be stable. Smith ([67], 1989) also investigated core-annular flows in vertical pipes taking gravity into account. The fluids have different densities and equal viscosities, and linear stability of the interface was explored using long-wavelength disturbances. He found that when the densities are similar the down flows are stable when the core fluid is lighter than the film fluid, and the upflows are stable when the core fluid is heavier than the film fluid. Russo and Steen ([62], 1989) studied the linear stability of an annular film flowing down a rod with a free surface. They conclude that if the film is thin enough the shear can stabilize capillary breakup. Hu and Joseph ([29], 1989) performed the linear stability analysis of the problem of concentric fluids with an oil film on the inside wall of the pipe and oil in the core with a lubricating layer of water between them. This is called the three layer problem. In addition, they considered an energy for the wave of maximum growth in order to analyze the instability.

In addition, Chen, Bai, and Joseph ([9], 1990) studied core-annular flows in vertical pipes considering gravity, and including all effects of viscosity stratification and surface tension. Georgiou, Maldarelli, Papageorgiou, and Rumschitzki ([21], 1992) analyzed the linear stability of a vertical, perfectly concentric core-annular flow in the limit when the film is much thinner than the core. Using asymptotic expansions, they developed a new theory for the linear stability of the wetting layer in low-capillary-number liquid-liquid displacements. Their results agree with the experimental results of Aul and Olbricht ([4], 1990). Joseph and Renardy ([38], 1993) discuss in detail the effects of interfacial tension, interfacial friction, and Reynolds stresses as mechanisms of instability. Huang and Joseph ([30], 1995) studied the effect of eccentricity on core-annular flows with matched densities. They conclude that the linear stability does not provide a stable center for core-annular flows in the case of matched densities.
Kouris and Tsamopoulos ([41], 2001) studied the linear stability of core-annular flow of two immiscible fluids in a periodically constricted tube (sinusoidally varying cross-section). Gravitational effects are neglected. Generally, steady core-annular flow in a straight tube is less susceptible to instability than in a periodically constricted tube. Some of their results are that at low values of the Reynolds number, the streamlines follow the tube walls and its symmetry, as expected under creeping flow conditions. Increasing the Reynolds number or decreasing the viscosity ratio breaks the symmetry. Recirculation is induced in the region of the tube with bigger cross-section.

**Nonlinear stability studies:** Dynamics of core-annular flows in which effects of nonlinearity are kept can be described by nonlinear stability theories. Some nonlinear theories are established on the supposition that the wavelength is long compared to some length scale, typically the pipe diameter or film thickness. Frenkel, Babchin, Levich, Shlang, and Sivashinsky ([19], 1987) studied two fluids in a straight tube with the annular one being thin. Both fluids have equal properties and only interfacial tension acts between them. They derived the Kuramoto-Sivashinsky equation which includes both stabilizing and destabilizing terms related to the interfacial tension, leading to growth of the initial disturbances. Frenkel ([18], 1988) considered the wavelength to be long compared to the annular thickness but of the order of the core radius. He developed a modified Kuramoto-Sivashinsky equation for slow flow and discussed how the extra terms in his equation could alter the behavior of the Kuramoto-Sivashinsky equation. Papageorgiou, Maldarelli, and Rumschitzky ([54], 1990) investigated the weakly nonlinear evolution of thin films (wavelength long compared to the annular thickness). They studied the core contribution by searching in a larger set of core flow regimes. They conclude that viscosity stratification greatly increases the likelihood of regular nonlinear traveling waves.
Kerchman ([39], 1995) modeled the problem of oil in the annular region using strongly nonlinear theory. A modified Kuramoto-Sivashinsky equation was derived with additional dispersive terms. By solving this last equation he found a large variety of solutions in the dynamics, from chaos to quasi-steady waves. Coward, Papageorgiou, and Smyrlis ([12], 1995) examined the case when the pressure gradient is modulated by time harmonic oscillations. Viscosity stratification and interfacial tension are present. They developed a weakly nonlinear asymptotic approximation valid for thin annular films. The spatio-temporal evolution equation for the interface is an extension of the equation derived for constant pressure gradient by Papageorgiou, Maldarelli, and Rumschitzky ([54], 1990) to a similar system with time periodic coefficients. Extensive numerical simulations are carried out and they conclude that the forcing promotes irregular oscillations in the presence of large dispersion which would otherwise produce traveling wave pulses in unforced flows.

Renardy ([61], 1997) studied core-annular flows of two fluids at the onset of a non-axisymmetric instability. She derived the weakly nonlinear amplitude evolution equations and found bifurcating solutions which are traveling waves and standing waves. The traveling waves move in the azimuthal and axial directions and are observed as corkscrew waves, see Figure 1.3(a). However, the standing waves travel only in the axial direction and are observed as snakes, see Figure 1.3(b).

Kouris and Tsamopoulos ([42], 2001) studied the nonlinear dynamics of a concentric, two-phase flow of immiscible fluids in a cylindrical tube. They extended their analysis and calculations in (Kouris and Tsamopoulos [40], 1999) when the more viscous fluid is in the core for any thickness of the film. Their results show that as the Reynolds number or the flow-rate of the core fluid increase, then the wavelength and the amplitude of the waves decrease. They obtained shapes in closer agreement with the observed ones in the experiments (Bai, Chen, and Joseph [5], 1992) than those in earlier investigations. Kouris and Tsamopoulos ([43], 2002) also studied the nonlinear
Figure 1.3  Core-annular flows of two fluids at the onset of a non-axisymmetric instability. For the case of traveling waves we observe (a) corkscrew waves, and for the case of standing waves we observe (b) snake waves. These figures are taken from Renardy ([61], 1997), Figure 1 (b) and (c).

dynamics of a concentric, two-phase flow of immiscible fluids in a cylindrical tube, with the less viscous fluid in the core (saw tooth waves).

1.2.3 Simulations for Clean Interfaces

Preziosi, Chen, and Joseph ([59], 1989) gave a careful numerical study of the case of a more viscous core for axisymmetric disturbances of all length scales in which all effects except gravity were included. They conclude that there is a window of stability when the annulus is thinner than a critical value and sufficiently less viscous than the core. As the viscosity of the annular film increases, this window disappears. Full numerical simulations of the Stokes flow; i.e., neglecting inertial effects in the flow, have been performed by Newhouse and Pozrikidis ([49], 1992). They simulate the flow evolution due to surface tension of an axisymmetric film coating the inner walls of a cylindrical tube. Newhouse and Pozrikidis assume that the fluids satisfy the creeping flow equation, so that a boundary integral numerical method can be employed. They
found that the ratio of the core radius to the cylinder radius has an influence on the volume of the drops. When that ratio is bigger than 0.82 the evolution of thin films leads to the formation of an array of collars.

Akrivis ([3], 1992) analyzed a Crank-Nicolson-type finite difference scheme for the Kuramoto-Sivashinsky equation in one space dimension with periodic boundary conditions. Papageorgiou and Smyrlis ([56], 1991) and Smyrlis and Papageorgiou ([68], 1991) developed extensive numerical simulations of the Kuramoto-Sivashinsky equation. They found different routes to chaos in which multimodal attractors and time periodic evolving to multiple period doublings sequentially appear. Later, a computational study of chaotic and ordered solutions of the Kuramoto-Sivashinsky equation was done by Smyrlis and Papageorgiou ([69], 1996). General initial conditions are used and evolving states do not assume odd-parity. They report on different routes to chaos and show that multimodal steady states emerge and are supported on decreasing windows in parameter space. Additionally, they present a self-similarity property of the Kuramoto-Sivashinsky equation showing that these profiles can be obtainable from global fixed point attractors of the equation at larger values of the “viscosity” parameter. They (Smyrlis and Papageorgiou [70], 1998) also present results from an extensive numerical experiments of the effects of dispersion on the Kuramoto-Sivashinsky equation and found that solutions are almost identical in both steady traveling waves and time periodic attractors.

Bai, Kelkar, and Joseph ([6], 1996) computed waves with constant bamboo wave forms assuming that the oil and water have the same densities. The viscosity of the core (oil) is relatively big and the waves are traveling downstream with a constant velocity. They partially succeed in computing waveforms with more rounded and asymmetric crests than the ones in (Bai, Chen, and Joseph [5], 1992).

Li and Renardy ([46], 1999) and Renardy and Li ([60], 2001) simulate numerically the flow of oil and water in an axisymmetric pipeline with a vertical core-annular
flow configuration. In their simulations they obtained bamboo type waves in down-flow and showed cases of waves behaving like the up-flow cases. Joseph and Renardy [38] is a good source for bamboo wave analysis and, in general, for core-annular flow analysis in the case of lubricating pipelining. Kouris and Tsamopoulos ([42], 2001 and [43], 2002) simulate the dynamics of two-phase flow of immiscible fluids in a circular pipe (bamboo and saw tooth waves, respectively), using pseudo-spectral numerical methods.

1.3 Theoretical Results with Surfactants

In general, the presence of even minute amounts of surfactant on a fluid-fluid interface can have a substantial effect on the evolution of the interface (Edwards, Brenner, and Wasan [16], 1991). Insoluble surfactants are large molecules possessing a dipolar structure formed of hydrophobic (i.e. water-repeling) and hydrophilic (i.e. water-attracting) segments; in this way, insoluble surfactants are distributed on interfaces separating aqueous and nonaqueous phases as water and oil. Surfactants influence the interfacial dynamics in two ways. Firstly, most types of surfactant reduce the interfacial tension, i.e. the surface tension in a surfactant coated interface is lower than that for a clean interface, with the interfacial tension correspondingly lower over regions of the interface with higher surfactant concentration. Secondly, the presence of a gradient in surfactant concentration introduces a Marangoni force. This is a force along the interface which is directed from regions of high surfactant concentration (i.e. low surface tension) to regions of low surfactant concentration (i.e. high surface tension). In general, the Marangoni force acts to oppose any external flow which promotes build-up or excess of surfactant along the interface. For a schematic representation of the Marangoni effects, see Figure 1.4.

The stability of jets in the presence of surfactants has been addressed by Whitaker ([81], 1976) who consider the temporal linear stability of initially quiescent
threads without any surrounding viscous fluid. With the presence of a surrounding fluid, Hansen, Peters and Meijer ([27], 1999), and Kwak and Pozrikidis ([44], 2001) have found that increasing the amount of surfactants initially present on the interface, decreases the growth rate and wavelength of the most unstable wave. Craster, Matar and Papageorgiou ([14], 2002) have studied several aspects of jet breakup and satellite formation in the presence of insoluble surfactants. They derived one-dimensional partial differential equations for the evolution of the interface and surfactant concentration in the long wavelength approximation. Numerical solutions show that the size of the satellite formed during breakup decreases with increasing initial surfactant concentration and surfactant activity.

There is a rather large literature devoted to numerical studies concerning the effect of surfactant on bubbles rising under gravity, in which we can mention Sadhal and Johnson ([64], 1983), Wang, Papageorgiou and Maldarelli ([78], 1999 and [79], 2002) and Wang ([77], 1999). Some theoretical studies of deforming drops under the effects of surfactants were developed by Flumerfelt ([17], 1980). He employed asymptotic analysis to investigate small deformations of spherical drops in extensional flows. Stone and Leal ([72], 1990), and Milliken and Leal ([47], 1994) repeated and
extended the calculation made by Flumerfelt ([17], 1980). Milliken, Stone, and Leal ([48], 1993) studied the evolution of drops under the effect of the viscosity ratio between the drop and the ambient fluid. Siegel ([66], 1999) employed a simple plane flow model to examine the deformation of a bubble in strain type flows and under the influence of surfactants.

Jensen and Halpern ([33], 1998) studied the stress singularity in surfactant-driven thin-film flows considering the effects of viscosity. They conclude that as surfactants increase in strength, they smooth the tip singularity over short length-scales, eliminate the local stress maximum and destroy the vortices. Also, Jensen ([32], 1998) studied the same problem but taking into account the inertial effects and established the conditions for which lubrication theory gives an accurate approximation for this type of surfactant-spreading flows.

The stability of core-annular flows in the presence of surfactant has received little attention. Most of the work on the effects of surfactant in core-annular flow have been motivated by applications to pulmonary fluid dynamics. The lung airways are internally coated by a thin film of a liquid forming a liquid-air interface. The interfacial tension tries to minimize the interfacial area. Thus, the coating liquid may cause closing off of the tiny airways by the formation of a meniscus during exhalation. Biological surfactant tends to reduce the interfacial tension by decreasing the attractive force between molecules of the film. A role of surfactant then, is to have a stabilizing effect which prevents collapses and keeps airways open. Otis, Johnson, Pedley, and Kamm ([52], 1990) numerically studied the effect of surfactant in annular liquid film collapse due to surface tension-driven instabilities. They found that surfactant can extend the life of an unstable film by at most a factor of four. Halpern and Grotberg ([24], 1992) considered the stability, including the effects of surfactant, of a liquid-lined flexible cylinder. In ([25], 1993), Halpern and Grotberg developed a nonlinear model taking into account the fluid mechanics of the thin film,
the equation of motion of the tube and the transport equation. They found that the convection of surfactant has the effect of reducing the surface tension which prevents meniscus formation. Otis, Johnson, Pedley, and Kamm ([53], 1993) developed a numerical model that simulates airway closure including the effects of both surfactant and time-varying geometry. They found that surfactant is effective in retarding or eliminating liquid bridging by reducing the mean surface tension and the action of surface gradients, and thus presumably reduces the tendency for airway compliant collapse. Motivated by the bilayer nature of fluid lining in the lungs' airways, Craster and Matar ([13], 2000) investigated surfactant transport on a bilayer fluid system. They derived a coupled spatio-temporal evolution equations describing the layer thickness and surfactant concentration.

Here, we want to explore the influence of surfactant in a core-annular flow when the core liquid is surrounded by another annular liquid. We assume the surfactant to be insoluble in the film and the core. This, physically, corresponds to surfactant that has a very low solubility in both the film and core fluids. So, the surfactant remains at the fluid-fluid interface.

In this work, we employ a long wave asymptotic analysis to carefully derive a coupled nonlinear system of equations. These equations, which govern the evolution of the interface and surfactant concentration, are solved numerically for a range of relevant system parameters in order to examine the influence of surfactants in the development of the interface, i.e. our goal is to analyze the effect of surfactant during the adjustment of the film. The nonlinear system derived is a forced Kuramoto-Sivashinky equation, the forcing arising from the Marangoni effect. We present extensive numerical experiments to quantify the large time dynamics. An implicit pseudo-spectral scheme which is spectral in space and second order finite difference in time is developed.
The rest of this work consists of five chapters and three appendices. Chapter 2 describes the mathematical model, governing equations, and details of the surface gradient operator required in our analysis. At the end of chapter 2 we give a summary of the dimensional model. Chapter 3 presents the asymptotic analysis leading to the evolution equations. Chapter 4 describes all the details regarding the implicit pseudo-spectral scheme, its implementation, and characterization of the attractors (solutions). In Chapter 5, we report the majority of the results obtained, and in Chapter 6, we give our conclusions. Appendix A gives the details regarding the curvature of the interface. In Appendix B, we give a proof of the $O((\Delta t)^2)$ accuracy of the scheme that we developed in Chapter 4, and finally, in Appendix C, we present diagnostics of the type of attractors (solutions).
CHAPTER 2

MATHEMATICAL MODEL AND GOVERNING EQUATIONS

Our problem consists of an annular liquid film (fluid 2), \(-\infty < z < \infty\), surrounding an infinitely long cylindrical fluid core (fluid 1) (see Figure 2.1). Fluid 1 is of undisturbed radius \(R_1\) and viscosity \(\mu_1\). The viscosity of fluid 2 is \(\mu_2\) and the tube is of radius \(R_2\). Here, we take the densities of the film and core fluids to be the same and equal to \(\rho\). Hence, gravitational effects are neglected (Hammond [26], 1983, Hu and Joseph [29], 1989, and Joseph and Renardy [38], 1993); gravity does not appreciably change the shape of the interface if the Bond number \(B_0 = \frac{\rho g a^2}{\sigma}\) is small. (The Bond number is the ratio between gravitational and capillary forces.) The flow is driven by a constant pressure gradient \(\nabla p = -F \epsilon_z\), where \(\epsilon_z = (0, 0, 1)\) and \(F > 0\). Insoluble surfactants are present on the fluid interface; we denote the surfactant concentration (in units of mass of surfactant per unit of interfacial area) by \(\Gamma^*\).

![Figure 2.1](image)

Figure 2.1 Core-annular flow of two immiscible fluids within a circular cylindrical pipe.
2.1 Governing Equations

The presence of surfactant decreases the interfacial tension. The relationship between the interfacial tension $\sigma$ and the surfactant concentration $\Gamma$ is given by the surface equation of state for the interfacial tension (Edwards, Brenner, and Wasan [16], 1991, Milliken, Stone, and Leal [48], 1993, and Stone and Leal [72], 1990)

$$\sigma \equiv \sigma(\Gamma) = \sigma_0 + 3RT\Gamma_\infty \ln (1 - \Gamma),$$  \hspace{1cm} (2.1)

where $\sigma_0$ is the interfacial tension of the clean (without surfactant) interface, $R$ is the ideal gas constant and $T$ is the temperature. The dimensionless surfactant concentration is given by $\Gamma = \frac{\Gamma}{\Gamma_\infty}$, where $\Gamma_\infty$ is the maximum packing concentration that the interface can support. Expanding $\ln (1 - \Gamma)$ in Taylor series about $\Gamma = 0$ we obtain the linear relation between the interfacial tension $\sigma$ and the surfactant concentration $\Gamma$ expressed as follows

$$\sigma(\Gamma) = \sigma_0(1 - \beta \Gamma),$$ \hspace{1cm} (2.2)

where $\beta = \frac{RgT\Gamma_\infty}{\sigma_0}$ and $\sigma_0$, $R$, $T$, $\Gamma_\infty$, and $\Gamma$ are defined as before. Expression (2.2) is expected to hold in the dilute $\Gamma$ limit. Even though this appears to be a restrictive assumption, our asymptotic solution is developed for small surfactant variations about a uniform state, in which instance (2.2) is the appropriate starting point.

We use cylindrical polar coordinates $\vec{x} = (r, \theta, z)$ with associated velocity components $\vec{u}_1 = (u_1, v_1, w_1)$ for the fluid core and $\vec{u}_2 = (u_2, v_2, w_2)$ for the fluid film, and we denote the interface between the fluids by $r = S(z, \theta, t)$. Our problem is axisymmetric; i.e., independent of the azimuthal variable $\theta$. The dimensional interface $S(z, t)$ can be written as

$$S(z, t) = R_1(1 + \delta H),$$ \hspace{1cm} (2.3)
where $R_1$ is the undisturbed core radius (see Figure 2.2), and $\delta$ is a dimensionless amplitude (see §3.3 and Figure 3.3).

**Figure 2.2** Half of a cross-section along the axis of a core-annular flow in a cylindrical pipe.

For the interface evolution, we start from the **Navier-Stokes** and **Continuity** equations for axisymmetric flows

\[
\rho (u_t + \vec{u} \cdot \nabla u) = -p_r + \mu \left( \nabla^2 u - \frac{u}{r^2} \right) \tag{2.4}
\]

\[
\rho (w_t + \vec{u} \cdot \nabla w) = -p_z + \mu \nabla^2 w \tag{2.5}
\]

\[
\nabla \cdot \vec{u} = 0 \tag{2.6}
\]

where $\mu$ is the fluid viscosity and $\rho$ the density. The gradient and divergence for axisymmetric flows, in cylindrical coordinates, are given by

\[
\nabla u = (u_r, 0, u_z) \tag{2.7}
\]

\[
\nabla \cdot \vec{u} = \frac{1}{r} (ru)_r + w_z \tag{2.8}
\]

respectively. Then,

\[
\rho (u_t + uu_r + uw_z) = -p_r + \mu \left( \nabla^2 u - \frac{u}{r^2} \right) \tag{2.9}
\]

\[
\rho (w_t + uw_r + ww_z) = -p_z + \mu \nabla^2 w \tag{2.10}
\]

\[
\frac{1}{r} (ru)_r + w_z = 0 \tag{2.11}
\]
with viscosity ($\mu_1$) and pressure ($p_1$) for the core, and viscosity ($\mu_2$) and pressure ($p_2$) for the film. The Laplacian ($\nabla^2$) in cylindrical coordinates for axisymmetric flows is defined as

$$\nabla^2 \equiv \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial}{\partial r} \right) + \frac{\partial^2}{\partial z^2}. \quad (2.12)$$

**Boundary Conditions**

In order to complete the mathematical model, we require a **no-slip condition** at the pipe wall $\bar{u}_2 = 0$, **continuity of velocity** at the interface $\bar{u}_1 = \bar{u}_2$, and the **kinematic condition** holding at the interface $r = S(z,t)$ (Joseph and Renardy [37], 1993),

$$u = S_t + w S_z. \quad (2.13)$$

We also require **Normal Stress Balance**

$$[\vec{n} \cdot \bar{T} \cdot \vec{n}]_1^2 = \sigma \nabla_s \cdot \vec{n}, \quad (2.14)$$

and **Tangential Stress Balance**

$$[\vec{t} \cdot \bar{T} \cdot \vec{n}]_1^2 = - \nabla_s \sigma \cdot \vec{t}, \quad (2.15)$$

where $[\cdot]^2_1 = (\cdot)_2 - (\cdot)_1$, $\bar{T}$ is the stress tensor, $\nabla_s$ is the surface gradient operator, $\vec{n}$ the normal unit vector pointing into region 1 (core), and $\vec{t}$ the tangential unit vector. For definitions of $\bar{T}$, $\nabla_s$, $\vec{n}$, and $\vec{t}$ see section §2.2 below.

For the surfactant concentration evolution, we start from the convective-diffusion equation for **surfactant transport** (Wong, Rumschitzki, and Maldarelli [82], 1996)

$$\frac{\partial \Gamma}{\partial t} - \frac{\partial \bar{x}}{\partial t} \cdot \nabla_s \Gamma + \nabla_s (\Gamma w_s) - D_s \nabla_s^2 \Gamma + \Gamma \kappa \bar{u} \cdot \vec{n} = 0, \quad (2.16)$$
where $\vec{x}$ is the position vector of the interface, $w_s$ is the surface velocity at the interface, $D_s$ is the constant interfacial surfactant diffusivity, and $\kappa$ is the curvature of the interface.

The various terms present in equations (2.14), (2.15), and (2.16), which are, $\vec{n} \cdot \vec{T} \cdot \vec{n}$, $\vec{t} \cdot \vec{T} \cdot \vec{n}$, $\nabla_s \cdot \vec{n}$, $\nabla_s \sigma \cdot \vec{t}$, $\frac{\partial \vec{x}}{\partial t} \cdot \nabla_s \Gamma$, $\nabla_s (\Gamma w_s)$, $\nabla_s^2 \Gamma$, and $\kappa$, are constructed in §2.2 next, using results from tensor analysis (Rutherford [63], 1989 and Wheeler and McFadden [80], 1994).

### 2.2 Surface Gradient Operator

In this section, we define the position vector, $\vec{x}$, in cylindrical coordinates and in terms of polar unit vectors. The interface is parametrized in terms of $\theta$ and $z$, that is we write $\vec{x} \equiv \vec{x}(\theta, z)$. It follows that any point on the axisymmetric interface has position vector

$$\vec{x} = S e_r + z e_z,$$

where $e_r = (\cos \theta, \sin \theta, 0)$, $e_\theta = (-\sin \theta, \cos \theta, 0)$, and $e_z = (0, 0, 1)$. A vector $u_s$ on the tangent plane to the surface $S$, is given by

$$u_s = u^1 \vec{t}_1 + u^2 \vec{t}_2,$$  \hfill (2.17)

where $\vec{t}_1$ and $\vec{t}_2$ are the contravariant basis vectors for $u_s$ at any point of the surface and $u^1$ and $u^2$ are the components of $u_s$ in the directions of $\vec{t}_1$ and $\vec{t}_2$, respectively. The vectors $\vec{t}_1$ and $\vec{t}_2$ are given by the partial derivatives of the position vector $\vec{x}$ as follows

$$\vec{t}_1 \equiv \vec{t}_1(\theta, z) = \frac{\partial \vec{x}}{\partial z} = S' e_r + e_z,$$

with $S' \equiv \frac{\partial S}{\partial z}$,

$$\vec{t}_2 \equiv \vec{t}_2(\theta, z) = \frac{\partial \vec{x}}{\partial \theta} = S e_\theta.$$
Therefore,

\[ u_s = \left(S'u^1, Su^2, u^1\right). \]

In order to find the normal unit vector, we introduce the covariant components, \( h_{11}, h_{12}, h_{21}, h_{22} \), (Wheeler and McFadden [80], 1994), of the surface metric tensor by

\[
\begin{align*}
    h_{11} &= \bar{t}_1 \cdot \bar{t}_1 = (S')^2 + 1 \\
    h_{12} &= h_{21} = \bar{t}_2 \cdot \bar{t}_1 = 0 \\
    h_{22} &= \bar{t}_2 \cdot \bar{t}_2 = S^2.
\end{align*}
\]

The determinant, \( h \), of the surface metric tensor can be defined in at least two ways. One way is using the covariant components, \( h_{11}, h_{12}, h_{21}, h_{22} \) of the surface tensor. Another way is using the square of the norm of the cross product \( \bar{t}_1 \times \bar{t}_2 \). The latter leads to

\[
h = h_{11}h_{22} - (h_{12})^2 = |\bar{t}_1 \times \bar{t}_2|^2 = S^2[(S')^2 + 1]
\]

Thus, we obtain the normal unit vector to the surface as

\[
\vec{n} = \frac{1}{\sqrt{h}} \bar{t}_1 \times \bar{t}_2 = \frac{1}{\sqrt{1 + (S')^2}} (S' \varepsilon_x - \varepsilon_x).
\]

The tangential unit vector to the surface, in the axial direction, is given by

\[
\vec{t} = \frac{\bar{t}_1}{|\bar{t}_1|} = \frac{1}{\sqrt{1 + (S')^2}} (S' \varepsilon_x + \varepsilon_x).
\]

For the purpose of calculating the surface gradient operator, \( \nabla_s \), we define the reciprocal basis vectors, \( \tilde{t}_1 \) and \( \tilde{t}_2 \), in terms of the covariant basis vectors, \( \bar{t}_1 \) and \( \bar{t}_2 \), as follows (Wheeler and McFadden [80], 1994)

\[
\tilde{t}_1 = -\frac{1}{\sqrt{h}} \vec{n} \times \bar{t}_2 = \frac{1}{h} \left( h_{22} \bar{t}_1 - h_{12} \bar{t}_2 \right).
\]
For our surface parametrization, the surface gradient operator, \( \nabla_s \), based on the reciprocal basis vectors \( \vec{t}^1 \) and \( \vec{t}^2 \), is given by

\[
\nabla_s = \vec{t}^1 \frac{\partial}{\partial z} + \vec{t}^2 \frac{\partial}{\partial \theta}.
\]

Therefore, in the calculation of \( \nabla_s \Gamma \), since \( \Gamma \) is a scalar function that does not depend on \( \theta \), the second term vanishes. Thus, the surface gradient of \( \Gamma \) is given by

\[
\nabla_s \Gamma = \vec{t} \frac{\partial \Gamma}{\partial z} = \frac{1}{1 + (S')^2} \frac{\partial \Gamma}{\partial z} (S', 0, 1).
\]

Let us express a vector \( \vec{V}(p, q) \) in its contravariant form

\[
\vec{V}(p, q) = V^1(p, q) \vec{t}^1 + V^2(p, q) \vec{t}^2 + V^3(p, q) \vec{n}.
\]

The divergence of \( \vec{V}(p, q) \) is given by

\[
\nabla_s \cdot \vec{V} = \frac{1}{\sqrt{h}} \left[ \frac{\partial (\sqrt{h} V^1)}{\partial p} + \frac{\partial (\sqrt{h} V^2)}{\partial q} \right] + V^3 \kappa,
\]

where \( \kappa \) is twice the mean curvature of the interface. We know, from (2.17), that \( u_s \Gamma = u^1 \Gamma \vec{t}^1 + u^2 \Gamma \vec{t}^2 \), thus the divergence of \( u_s \Gamma \) is given by

\[
\nabla_s \cdot (u_s \Gamma) = \frac{1}{\sqrt{h}} \left[ \frac{\partial (\sqrt{h} u^1 \Gamma)}{\partial z} + \frac{\partial (\sqrt{h} u^2 \Gamma)}{\partial \theta} \right],
\]

but our problem is independent of \( \theta \), so the second term in (2.24) vanishes. Taking the dot product of \( u_s \Gamma \) with \( \vec{t}^1 \), we obtain

\[
u^1 \Gamma = \frac{\Gamma}{1 + (S')^2} u_s \cdot \vec{t}^1 = \frac{\Gamma}{1 + (S')^2} (S' \xi_p + \xi_x) = \frac{\Gamma}{1 + (S')^2} (w + S' u),
\]
casting (2.24) into

\[ \nabla_s \cdot (u_s \Gamma) = \frac{1}{S \sqrt{1 + (S')^2}} \left[ \frac{\partial}{\partial z} \left( \frac{S T}{\sqrt{1 + (S')^2}} (w + S'u) \right) \right]. \]

We also have,

\[ \frac{\partial \vec{r}}{\partial t} = \frac{\partial \vec{S}}{\partial t} \cdot \vec{r}, \]

which combined with (2.22), yields the result

\[ \frac{\partial \vec{r}}{\partial t} \cdot \nabla_s \Gamma = \frac{\partial \Gamma}{\partial z} \frac{1}{1 + (S')^2} \left( \frac{\partial \vec{S}}{\partial t} \cdot 0, 0 \right) \cdot (S', 0, 1) = S' \frac{\partial \vec{S}}{\partial t} \frac{\partial \Gamma}{\partial z} \frac{1}{1 + (S')^2}. \]

In what follows, we calculate \( \nabla_s^2 \Gamma \). We are able to do so by using the definitions of surface gradient and surface divergence defined above. Consequently,

\begin{align*}
\nabla_s^2 \Gamma &\equiv \nabla_s \cdot (\nabla_s (\Gamma)) = \nabla_s \cdot (\nabla_s \Gamma) \\
&= \nabla_s \cdot \left( \frac{1}{1 + (S')^2} \frac{\partial \Gamma}{\partial z} (S', 0, 1) \right) \\
&= \nabla_s \cdot \left( \frac{1}{1 + (S')^2} \frac{\partial \Gamma}{\partial z} \hat{r}_1 \right). \quad (2.26)
\end{align*}

and applying the definition of divergence from (2.23), we finally obtain an expression for the surface Laplacian, \( \nabla_s^2 \), of the concentration of surfactant \( \Gamma \), as

\[ \nabla_s^2 \Gamma = \frac{1}{S \sqrt{1 + (S')^2}} \frac{\partial}{\partial z} \left( \frac{S}{\sqrt{1 + (S')^2}} \frac{\partial \Gamma}{\partial z} \right). \quad (2.27) \]

The curvature, \( \kappa \), is the surface divergence of the normal unit vector, i.e. \( \kappa = \nabla_s \cdot \hat{n} \); but also, the curvature is equivalently given by \( \kappa = \kappa_1 + \kappa_2 \), where \( \kappa_1 = \frac{1}{R_1} \) and \( \kappa_2 = \frac{1}{R_2} \) with \( R_1 \) the radius of the circumference circumscript to the interface in a plane parallel to the interfacial normal vector, and \( R_2 \) the radius of the circumference circumscript to the interface in a plane parallel to the cylinder axis. For an explanation of \( \kappa_1 \) and \( \kappa_2 \), as given below, see Appendix A. Thus,

\[ \kappa_1 = \frac{S''}{(1 + (S')^2)^{3/2}} \quad (2.28) \]
Hence, we can obtain the last term in the surfactant transport equation as a function of the interface variable, $S$, and velocity components, $u$, and $w$.

$$
\kappa u \cdot \vec{n} = \frac{1}{S(1 + (S'))} \left[ 1 - \frac{SS''}{1 + (S')^2} \right] (-S'u + v).
$$

We still need to calculate the left hand side of the normal stress balance, $\vec{n} \cdot T \cdot \vec{n}$, and the left hand side of the tangential stress balance, $F \cdot T \cdot \vec{n}$. We start with the stress tensor, $\underline{T} \equiv T_{ij}$ (Batchelor [7], 1999),

$$
T_{ij} \equiv \tau_{ij} = -p\delta_{ij} + 2\mu e_{ij},
$$

where $e_{ij}$ in the axisymmetric geometry are defined by

$$
e_{11} = e_{rr} = \frac{\partial u}{\partial r}, \quad e_{22} = e_{\theta\theta} = \frac{w}{z}, \quad e_{33} = e_{zz} = \frac{\partial w}{\partial z}
$$

$$
e_{12} = e_{r\theta} = 0, \quad e_{23} = e_{\theta z} = 0, \quad e_{31} = e_{zr} = \frac{1}{2} \frac{\partial u}{\partial z} + \frac{1}{2} \frac{\partial w}{\partial r},
$$

where $e_{ij} = e_{ji}$. The projection of all $[e_{ij}]$ onto the normal vector, $\vec{n}$, can be expressed in the following way

$$
[e_{ij}] \cdot \vec{n} = \frac{1}{\sqrt{1 + (S')^2}} \left( S' \frac{1}{2} \left( \frac{\partial u}{\partial z} + \frac{\partial w}{\partial r} \right) - \frac{\partial u}{\partial r}, 0, S' \frac{\partial w}{\partial z} - \frac{1}{2} \left( \frac{\partial u}{\partial z} + \frac{\partial w}{\partial r} \right) \right)
$$

and $[\delta_{ij}]$ projected onto the normal vector, $\vec{n}$, is just the normal vector, so $[\delta_{ij}] \cdot \vec{n} = \vec{n}$. Hence, the projection of the tensor, $\underline{T}$, onto the normal vector, $\vec{n}$, can be explicitly written as

$$
\underline{T} \cdot \vec{n} = -p\vec{n} + \frac{2\mu}{\sqrt{1 + (S')^2}} \left( S' \frac{1}{2} \left( \frac{\partial u}{\partial z} + \frac{\partial w}{\partial r} \right) - \frac{\partial u}{\partial r}, 0, S' \frac{\partial w}{\partial z} - \frac{1}{2} \left( \frac{\partial u}{\partial z} + \frac{\partial w}{\partial r} \right) \right).
$$
Finally, using the expression for the curvature (2.30) and the definition of the surface gradient (2.22), we have

\[
\sigma(\Gamma) \nabla_s \cdot \vec{n} = \frac{\sigma(\Gamma)}{S\sqrt{1 + (S')^2}} \left\{ 1 - \frac{SS''}{1 + (S')^2} \right\}
\]

\[-\nabla_s \sigma \cdot \vec{t} = \left[ -\frac{1}{1 + (S')^2} \frac{\partial \sigma}{\partial z} (S', 0, 1) \right] \cdot \left[ \frac{-1}{\sqrt{1 + (S')^2}} (S', 0, 1) \right]
\]

\[= \frac{1}{\sqrt{1 + (S')^2}} \frac{\partial \sigma}{\partial z},\]

respectively. Using these developments we obtain that the **Normal Stress Balance** is

\[
\left\{ -p + \frac{2\mu_i}{1 + (S')^2} \left[ (S')^2 \frac{\partial w}{\partial z} - S' \left( \frac{\partial u}{\partial z} + \frac{\partial w}{\partial r} \right) - \frac{\partial u}{\partial r} \right] \right\}^2 = \frac{\sigma(\Gamma)}{S\sqrt{1 + (S')^2}} \left\{ 1 - \frac{SS''}{1 + (S')^2} \right\},
\]

where \( i = 1, 2 \).

The **Tangential Stress Balance** is

\[
\left\{ \frac{\mu_i}{1 + (S')^2} \left[ 2S' \left( \frac{\partial u}{\partial r} - \frac{\partial w}{\partial z} \right) + (1 - (S')^2) \left( \frac{\partial u}{\partial z} + \frac{\partial w}{\partial r} \right) \right] \right\}^2 = \frac{-1}{\sqrt{1 + (S')^2}} \frac{\partial \sigma}{\partial z},
\]

where \( i = 1, 2 \).
The convective-diffusion equation for surfactant transport is

\[
\frac{\partial \Gamma}{\partial t} - \frac{\dot{S}S'}{1 + (S')^2} \frac{\partial \Gamma}{\partial z} + \frac{1}{S\sqrt{1 + (S')^2}} \left\{ \frac{\partial}{\partial z} \left[ \frac{S\Gamma}{\sqrt{1 + (S')^2}}(w + S'u) \right] \right\} \\
- \frac{D_s}{S\sqrt{1 + (S')^2}} \frac{1}{\partial z} \left( \frac{S}{\sqrt{1 + (S')^2}} \frac{\partial \Gamma}{\partial z} \right)
\]

\[+ \frac{\Gamma}{S(1 + (S')^2)} \left[ 1 - \frac{SS''}{1 + (S')^2} \right] (-S'w + u) = 0. \tag{2.38}
\]

### 2.3 Summary of the Dimensional Mathematical Model

In this section, we give all the governing equations and boundary conditions of our dimensional mathematical model that we developed in sections §2.1 and §2.2.

#### 2.3.1 Governing Equations

- **Navier-Stokes equations and incompressibility condition:**

  \[
  \rho(u_t + uu_r + uw_z) = -p_r + \mu \left( \nabla^2 u - \frac{u}{r^2} \right) \\
  \rho(w_t + uw_r + ww_z) = -p_z + \mu \nabla^2 w \\
  \frac{1}{r}(ru)_r + w_z = 0,
  \]

- **Convective-diffusion equation for surfactant transport:**

  \[
  \frac{\partial \Gamma}{\partial t} - \frac{\dot{S}S'}{1 + (S')^2} \frac{\partial \Gamma}{\partial z} + \frac{1}{S\sqrt{1 + (S')^2}} \left\{ \frac{\partial}{\partial z} \left[ \frac{S\Gamma}{\sqrt{1 + (S')^2}}(w + S'u) \right] \right\} \\
  - \frac{D_s}{S\sqrt{1 + (S')^2}} \frac{1}{\partial z} \left( \frac{S}{\sqrt{1 + (S')^2}} \frac{\partial \Gamma}{\partial z} \right)
  \]

  \[+ \frac{\Gamma}{S(1 + (S')^2)} \left[ 1 - \frac{SS''}{1 + (S')^2} \right] (-S'w + u) = 0. \]
2.3.2 Boundary Conditions

• No slip at the pipe wall and continuity of velocity at the interface:

\[
\vec{u}_2 = 0 \quad \text{at } r = R_2 \quad \text{and} \quad \vec{u}_1 = \vec{u}_2, \quad \text{at } r = S(t, z) \tag{2.39}
\]

• Kinematic condition:

\[
u = S_t + wS_z, \quad \text{at the interface } r = S(t, z). \tag{2.40}
\]

• Normal stress balance:

\[
\left\{ -p + \frac{2\mu_i}{1 + (S')^2} \left[ (S')^2 \frac{\partial w}{\partial z} - S' \left( \frac{\partial u}{\partial z} + \frac{\partial w}{\partial r} \right) + \frac{\partial u}{\partial r} \right] \right\}_1^2 = \frac{\sigma(\Gamma)}{S\sqrt{1 + (S')^2}} \left\{ 1 - \frac{SS''}{1 + (S')^2} \right\},
\]

where \(i = 1, 2\).

• Tangential stress balance:

\[
\left\{ \frac{\mu_i}{1 + (S')^2} \left[ 2S' \left( \frac{\partial u}{\partial r} - \frac{\partial w}{\partial z} \right) + (1 - (S')^2) \left( \frac{\partial u}{\partial z} + \frac{\partial w}{\partial r} \right) \right] \right\}_1^2 = \frac{-1}{\sqrt{1 + (S')^2}} \frac{\partial \sigma}{\partial z},
\]

where \(i = 1, 2\).
CHAPTER 3

DERIVATION OF EVOLUTION EQUATIONS

3.1 Non-dimensionalization

In order to non-dimensionalize lengths, we select the base core radius $R_1$. Velocities are non-dimensionalized by the centerline velocity $W_0$, time by $R_1/W_0$, interfacial tension by the surface tension $\sigma_0$ in the absence of surfactants, which is called the 'clean' surface tension, and pressure by $\rho W_0^2$, where $\rho$ is the density of the fluids.

Next, we write the nondimensional equations of our model. For the Navier-Stokes equations the nondimensionalization introduces the Reynolds numbers $(Re_i)$, $i = 1$ for the core fluid and $i = 2$ for the film fluid, defined by $Re_i = \rho W_0 R_1 / \mu_i$ corresponding to the relative importance of the inertial and viscous forces acting on unit volume of the fluid $i$. The nondimensionalization of the surfactant transport equation produces the Peclet number $(Pe)$ which defines the transport ratio between convection and diffusion and is given by $Pe = \frac{W_0 R_1}{D_s}$. In the normal stress balance, the nondimensionalization leads to a surface tension parameter $J = \frac{\sigma_0 R_1}{\rho \nu_1^2}$. The Capillary number $(C_a)$ and viscosity ratio $m$ arise in the dimensionless tangential stress balance. The capillary number is given by $C_a = \frac{\mu_i W_0}{\sigma_0}$. It measures the relative ratio between the base flow velocity and the capillary velocity. The viscosity ratio is given by $m = \frac{\mu_2}{\mu_1}$, the ratio of the film fluid viscosity to the core fluid vistosity. Note that the capillary number can be expressed in terms of $Re_1$ and $J$ as $C_a = \frac{Re_1}{J}$ and the viscosity ratio in terms of $Re_1$ and $Re_2$ as $m = \frac{Re_1}{Re_2}$.

We will use the same notation for dimensional and non-dimensional variables. The dimensionless Navier-Stokes equations and the continuity equation are

\begin{align}
(u_i)_t + u_i(u_i)_r + w_i(u_i)_z &= -(p_i)_r + \frac{1}{Re_i} \left[ \nabla^2 u_i - \frac{u_i}{r^2} \right], \\
(w_i)_t + u_i(w_i)_r + w_i(w_i)_z &= -(p_i)_z + \frac{1}{Re_i} \nabla^2 w_i,
\end{align}

(3.1) (3.2)
\[(u_i)_r + \frac{1}{r}u_i + (w_i)_z = 0, \quad (3.3)\]

where \(i = 1, 2\) for core and film respectively. The dimensionless surfactant equation is

\[
\frac{\partial \Gamma}{\partial t} - \frac{\dot{S}S'}{1 + (S')^2} \frac{\partial \Gamma}{\partial z} + \frac{1}{S \sqrt{1 + (S')^2}} \left\{ \frac{\partial}{\partial z} \left[ \frac{ST}{\sqrt{1 + (S')^2}} (w + S'u) \right] \right\} - \frac{1}{P_e} \frac{1}{S \sqrt{1 + (S')^2}} \frac{\partial}{\partial z} \left( \frac{S}{\sqrt{1 + (S')^2}} \frac{\partial \Gamma}{\partial z} \right) + \frac{\Gamma}{S(1 + (S')^2)} \left[ 1 - \frac{SS''}{1 + (S')^2} \right] (S'w + u) = 0. \quad (3.4)\]

The no-slip condition at the pipe wall is

\[u_2 = w_2 = 0 \quad \text{at} \quad r = \frac{R_2}{R_1}, \quad (3.5)\]

the continuity of velocities is

\[\{u_i\}_1^2 = 0, \quad \{w_i\}_1^2 = 0 \quad \text{on} \quad r = S(z, t),\]

and the Kinematic condition is

\[u = \frac{\partial S}{\partial t} + w \frac{\partial S}{\partial z} = S_t + wS'.\]

The dimensionless normal stress balance is

\[
\left\{ p(1 + (S')^2) - \frac{2}{Re_t} \left[ (S')^2 w_z - S' (u_z + w_r) + u_r \right] \right\}_1^2 = \frac{J(1 - \beta \Gamma)}{Re_t^2} \left\{ S'' - \frac{1}{S} \left[ 1 + (S')^2 \right] \right\} \left[ 1 + (S')^2 \right]^{-\frac{1}{2}}, \quad (3.6)\]

and the dimensionless tangential stress balance is

\[
\left\{ m_i \left[ 2S' (u_r - w_z) + [1 - (S')^2] (u_z + w_r) \right] \right\}_1^2 = \frac{\beta \Gamma}{C_a} \left[ 1 + (S')^2 \right]^{\frac{1}{2}}, \quad (3.8)\]
where \( m_1 = 1 \) and \( m_2 = m \).

### 3.2 Base State Solution

We begin our stability analysis by finding the dimensionless basic state driven by a constant pressure gradient \( p_z = -\frac{FR_1}{\rho W_0^2} \). This means that \( u_i = 0 \), \( w_i = 0 \), \( \Gamma = \Gamma_0 \) a constant with \( 0 < \Gamma_0 < 1 \), and \( S = 1 \). From the equation (3.1) we obtain \( 0 = -p_i \), so \( p_i \) is not a function of \( r \). From the continuity equation (3.3) \( w_i = 0 \). Thus, the Navier-Stokes equation (3.2) and the tangential stress balance (3.9) become

\[
p_i = \frac{1}{Re_i} \nabla^2 w_i,
\]

and

\[
Re_2 w_1 \mid_{r=1} = Re_1 w_2 \mid_{r=1},
\]

respectively. The surfactant equation (3.4) is automatically satisfied.

The equation (3.10) becomes

\[
-\frac{FR_1}{\rho W_0^2} Re_i = \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial w_i}{\partial r} \right)
\]

Multiplying the last equation by \( r \) and integrating it with respect to \( r \), we obtain

\[
r \frac{\partial w_i}{\partial r} = -\frac{1}{2} \frac{FR_1}{\rho W_0^2} Re_i r^2 + C_i.
\]

Dividing again by \( r \) and integrating with respect to \( r \), we obtain

\[
w_i = -\frac{1}{4} \frac{FR_1}{\rho W_0^2} Re_i r^2 + C_i \log r + D_i,
\]

where \( i = 1 \) is for the core and \( i = 2 \) is for the film.

Since \( w_1 \) must be bounded as we approach the tube axis \( (r = 0) \), \( C_1 = 0 \). From the tangential stress balance (3.2), we find \( C_2 = 0 \). The no slip condition at the pipe
wall, \( w_2\mid_{r=a} = 0 \), where \( a = R_2/R_1 \) gives

\[
D_2 = \frac{1}{4} \frac{FR_1}{\rho W_0^2} Re_2 a^2.
\]

Continuity of axial velocity at the interface, \( w_1\mid_{r=1} = w_2\mid_{r=1} \), implies

\[
D_1 = \frac{1}{4} \frac{FR_1}{\rho W_0^2} Re_1 - \frac{1}{4} \frac{FR_1}{\rho W_0^2} Re_2 (1 - a^2)
\]

Thus,

\[
w_1 = -\frac{1}{4} \frac{FR_1}{\rho W_0^2} Re_1 (r^2 - 1) - \frac{1}{4} \frac{FR_1}{\rho W_0^2} Re_2 (1 - a^2), \tag{3.11}
\]

\[
w_2 = -\frac{1}{4} \frac{FR_1}{\rho W_0^2} Re_2 (r^2 - a^2). \tag{3.12}
\]

Using the definition of the Reynolds number, \( Re_i = \frac{\rho w_0 R_1}{\mu_i} \), we can rewrite \( w_1 \) and \( w_2 \) as

\[
w_1 = -\frac{1}{4\mu_1} \frac{FR_1^2}{W_0} (r^2 - 1) - \frac{1}{4\mu_2} \frac{FR_1^2}{W_0} (1 - a^2), \tag{3.13}
\]

\[
w_2 = -\frac{1}{4\mu_2} \frac{FR_1^2}{W_0} (r^2 - a^2). \tag{3.14}
\]

Let us find an expression for \( W_0 \), the centerline velocity. We know that the dimensionless centerline velocity is \( w_1(r = 0) = 1 \), then

\[
1 = \frac{1}{4\mu_1} \frac{FR_1^2}{W_0} - \frac{1}{4\mu_2} \frac{FR_1^2}{W_0} \left(1 - \frac{R_2^2}{R_1^2}\right).
\]

Solving for \( W_0 \), the dimensional centerline velocity, we obtain

\[
W_0 = \frac{1}{4\mu_1\mu_2} [(\mu_2 - \mu_1)R_1^2 + \mu_1 R_2^2].
\]

Let us substitute \( W_0 \) in (3.13) and (3.14) to express \( w_1 \) and \( w_2 \) in closed form in terms of \( r, a, \) and \( m \)

\[
w_1 = 1 - \frac{mr^2}{a^2 + m - 1}, \quad 0 \leq r \leq 1, \tag{3.15}
\]
The difference in pressures can be found from the normal stress balance (3.7). Thus, the difference in pressures of the basic core-annular flow is

$$p_2 - p_1 = -\frac{J(1 - \beta \Gamma_0)}{Re_t^2} = -\frac{\sigma_0(1 - \beta \Gamma_0)}{\rho W_0^2 R_t},$$  \hspace{1cm} (3.17)

where $p_1$ and $p_2$ are the basic core and film pressures, respectively.

### 3.3 Derivation

In this section, we develop an asymptotic solution when the thickness of the film (relative to the core) is small, and derive a coupled system of leading order evolution equations. One equation describes the spatio-temporal evolution of the free interface between the core and the film, and the other describes the evolution of the concentration of surfactant. In the **nondimensional undisturbed state**, the core radius is 1 and the distance from the wall to the interface is $\varepsilon = a - 1$, where $a = R_2/R_1$. See Figure 3.3 for a better visualization of the situation. We proceed asymptotically with $\varepsilon << 1$. Let the interface deform to heights of order $\delta$, where $\delta << \varepsilon$. (This corresponds to a weakly nonlinear theory; in the absence of a background flow, we
can deal with $\delta \sim \varepsilon$.) The disturbed interface, then, is given by $S(z, t) = 1 + \delta H(z, t)$; this implies a dimensionless film thickness $\varepsilon - \delta H(z, t)$.

A local variable is introduced in the film region by $r = a - \varepsilon y$, where $y$ is 0 at the pipe wall and $1 - \frac{\delta}{\varepsilon} H(z, t)$ at the interface. The axial component is defined as $z = \dot{z}$. With the new scales, we have

\[
\begin{align*}
\frac{\partial}{\partial r} & \rightarrow \frac{1}{\varepsilon} \frac{\partial}{\partial y} \\
\frac{\partial}{\partial z} & \rightarrow \frac{\partial}{\partial \dot{z}}
\end{align*}
\] (3.18) (3.19)

and for simplicity, we drop the symbol.

### 3.3.1 Derivation of the Interface Evolution Equation

The scaled continuity equation is

\[
\frac{\partial w_2}{\partial z} - \frac{1}{\varepsilon(a - \varepsilon y)} \frac{\partial}{\partial y} ((a - \varepsilon y) u_2) = 0,
\] (3.20)

where $a = R_2/R_1 = 1 + \varepsilon$. Balance of terms for small $\varepsilon$ provides an estimate for $u_2$ to be equal to $\varepsilon$ times the order of $w_2$. In a similar way, from the continuity equation in the core, $u_1$ and $w_1$ must be of the same order.

Suppose $p_1 = \tilde{p}_1 + \hat{p}_1$ and $p_2 = \tilde{p}_2 + \hat{p}_2$, where $\tilde{\phantom{p}}$ and $\hat{\phantom{p}}$ indicate base and perturbed states, respectively. From the Navier-Stokes equation (3.2) in the film and supposing $(\tilde{p}_2)_z \sim 1$, we find that balancing viscous terms with the perturbation pressure gradient (this is essentially a lubrication approximation) gives

\[
\tilde{p}_{2z} \sim \frac{1}{Re_2 \varepsilon^2} \text{(order of } w_2) .
\]

Similarly, from the Navier-Stokes equation (3.2) in the core and using $(\tilde{p}_1)_z \sim 1$, we find that

\[
\tilde{p}_{1z} \sim \frac{1}{Re_1} \text{(order of } w_1) .
\]
Continuity of axial velocity at the interface yield order of \( w_1 = \text{order of } w_2 \). Therefore,

\[
\text{order of } \tilde{p}_1 = \frac{1}{Re_1} \text{ order of } w_2 \ll \frac{1}{m \epsilon^2 Re_2} \text{ order of } w_2 \sim \text{ order of } \tilde{p}_2, \quad (3.21)
\]

where we considered \( m \sim O(1) \). Considering the normal stress balance (3.7), we obtain the order of the perturbation pressure to be \( \tilde{p}_2 \sim \frac{J \delta}{Re_1^2} \). This scaling reflects the fact that we are considering capillary driven motions which arise from pressure changes due to the presence of surface tension. Therefore,

\[
w_2 \sim \frac{\epsilon^2 \delta J Re_2}{Re_1^2} \quad \text{and} \quad u_2 \sim \frac{\epsilon^3 \delta J Re_2}{Re_1^2}.
\]

For the case of \( m = \frac{Re_1}{Re_2} \sim O(1) \), the film perturbation velocities can be estimated to be

\[
w_2 \sim \frac{\epsilon^2 \delta J}{Re_1} \quad \text{and} \quad u_2 \sim \frac{\epsilon^3 \delta J}{Re_1}. \quad (3.22)
\]

Evaluating (3.15) and (3.16) the basic axial velocities, \( w_1(r) \) and \( w_2(r) \), at the disturbed interface, \( r = S = 1 + \delta H \), we obtain the difference in basic axial velocity, \( w_1 - w_2 \), as follows

\[
(w_1 - w_2)|_{r=1+\delta H} = \frac{2(1 - m) \delta H + O(\delta^2)}{m + 2 \epsilon - \epsilon^2} \sim O(\delta). \quad (3.23)
\]

Physically, this arises from the difference in viscosities or what is known as viscosity stratification. We already know, from (3.22), that the axial velocity perturbation in the film is of \( O\left(\frac{\epsilon^2 \delta J}{Re_1}\right) \), implying that the core contribution must be of \( O(\delta) \), to ensure continuity of velocities.

Considering the tangential stress balance (3.9)

\[
m \left[ 2S_z(u_{2r} - w_{2s}) + [1 - (S_z)^2](u_{2z} + w_{2s}) \right] - \\
\left[ 2S_z(u_{1r} - w_{1s}) + [1 - (S_z)^2](u_{1z} + w_{1s}) \right] = -\frac{\beta}{C_a} \Gamma_z [1 + (S_z)^2]^{\frac{1}{2}},
\]
we see that the dominant term in the film is the radial derivative of the axial film velocity, \( w_2 \), which is of \( O\left( \frac{\varepsilon J}{Re_1} \right) \) and the core contribution is of \( O(\delta) \). So, we have to consider various regimes.

**Regime 1. Film and core do not couple (film contribution dominates over the core contribution):**

If the film contribution dominates over the core contribution; i.e. \( \varepsilon J >> Re_1 \) and \( C_a << \varepsilon \), then the core influence is not introduced into the dynamics of the problem to leading order. This decoupling is a result of the core contribution in the tangential stress balance equation (3.9) being of lower order than the corresponding film contribution. The kinematic condition (3.1)

\[
\mathbf{u} = S_t + (\mathbf{w} + \mathbf{w})S_z,
\]

where \( \mathbf{w} \) represents the base state axial velocity and \( \mathbf{w} \) represents the perturbed axial velocity, taken in a frame of reference traveling with speed \( \mathbf{w}(r = 1; \varepsilon) \sim O(\varepsilon) \), provides an estimate for \( \delta \), the interfacial amplitude. This comes from balancing \( \mathbf{u} \) and the convective term on the right hand side, as well as allowing for unsteadiness on a new long time scale. We find

\[
\delta = \frac{\varepsilon J}{Re_1} >> \varepsilon^2,
\]

since \( \frac{\varepsilon J}{Re_1} >> 1 \). The size of \( \delta \) depends on the magnitude of \( \frac{\varepsilon J}{Re_1} \) and the evolution ranges from highly nonlinear regimes, \( \delta \sim \varepsilon \), leading to Hammond type equations (Hammond [26], 1983), or weakly nonlinear regimes leading to the Kuramoto-Sivashinsky equation (Smyrlis and Papageorgiou [68], 1991 ;[69], 1996 and [70], 1998).

We are interested in the case when film and core couple, and in what follows, we describe these delicate scalings in full detail.
Regime 2. Film and core couple:

If the film contribution and core contribution balance, then $\varepsilon J \sim \text{Re}_1$ and $C_a \sim \varepsilon$. In this regime, we consider two cases. One case with moderate surface tension ($J \sim O(1)$) and slow moving core ($\text{Re}_1 \sim O(1)$). Another case with strong surface tension ($J \sim O(1/\varepsilon)$) and moderate core flow ($\text{Re}_1 \sim O(1)$).

Let us consider the kinematic condition using the film variables

$$u_2 = S_t + (\bar{w}_2 + \bar{w}_2)S_z,$$  \hspace{1cm} (3.24)

where $\bar{w}_2$ and $\bar{w}_2$ represent the base state and perturbed axial velocities of the film, respectively. The term $u_2$ is of $O(\varepsilon^2 \delta)$, $S_t$ is of order $\delta$ times order of the time scale $t$, and the term $(\bar{w}_2 + \bar{w}_2)S_z$ is of $O(\varepsilon \delta)$, and therefore, we can not balance them.

In order to overcome this problem, we define a system of coordinates traveling with speed $\bar{w}_2$, where $\bar{w}_2|_{t=1+\delta t} = \bar{w}_2 + \bar{w}_2$, with $\bar{w}_2 \sim O(\varepsilon)$ and $\bar{w}_2 \sim O(\delta)$. This is called the Galilean transformation which we express as

$$\frac{\partial}{\partial t} \rightarrow -\bar{w}_2 \frac{\partial}{\partial z} + \frac{\partial}{\partial \tau}.$$  \hspace{1cm} (3.25)

So, plugging (3.25) in the kinematic condition (3.24) and dropping the ~ symbol, we obtain

$$u_2 = -\bar{w}_2S_z + S_t + (\bar{w}_2 + \bar{w}_2)S_z$$

$$= S_t + (\bar{w}_2 + \bar{w}_2)S_z,$$

and the term $u_2$ is of $O(\varepsilon^2 \delta)$, $S_t$ is of order $\delta$ times order of the time scale $t$, and the term $(\bar{w}_2 + \bar{w}_2)S_z$ is of $O(\delta^2)$. Balancing all the terms and introducing a new time variable $\tau$ we conclude that $\delta = \varepsilon^2$ and $\tau = \delta t$.

Consider the case $J \sim O(1)$ and $\text{Re}_1 \sim O(\varepsilon)$ of the regime 2. Then, in the film
\( \begin{aligned}
\varepsilon^4 \bar{u}_2 &= O(\varepsilon^5) \\
\varepsilon^3 \bar{\bar{w}}_2 &= O(\varepsilon^4) \\
\bar{p}_2 &= \bar{\bar{p}}_0 + \varepsilon \bar{p}_2 + \ldots
\end{aligned} \) (3.26)

and in the core

\( \begin{aligned}
\varepsilon^2 \bar{u}_1 &= O(\varepsilon^3) \\
\bar{w}_1 &= \bar{\bar{w}}_1 + \varepsilon \bar{\bar{w}}_1 + O(\varepsilon^3) \\
\bar{p}_1 &= \bar{\bar{p}}_1 + \varepsilon \bar{\bar{p}}_1 + \ldots
\end{aligned} \) (3.29)

Set \( Re_1 = \lambda \varepsilon \), where \( \lambda \sim O(1) \). Substituting \( u_2, w_2 \) and \( p_2 \) into the Navier-Stokes equations and using \( Re_1 = m Re_2 \), we obtain to leading order from (3.2)

\( \bar{\bar{p}}_0 + \frac{m}{\lambda} \bar{\bar{w}}_{2y} = 0. \) (3.32)

From (3.1) to leading order, we obtain \( \bar{\bar{p}}_{0y} = 0 \) while the continuity equation (3.3) gives \( \bar{\bar{w}}_{2z} = \bar{\bar{w}}_{2y} \), to leading order.

Since \( \bar{\bar{p}}_0 \) is not a function of \( y \), we integrate (3.32) twice to get

\( \bar{\bar{w}}_2 = \frac{\lambda}{m} \left( \frac{1}{2} \bar{\bar{p}}_0 y^2 + A(z, t)y \right) \),

where the no-slip condition \( \bar{\bar{w}}_2(y = 0, z, t) = 0 \) has been used. In order to find \( \bar{\bar{u}}_2 \), we differentiate \( \bar{\bar{w}}_2 \) with respect to \( z \) and then integrate the resulting equation over \( y \). Thus,

\( \bar{\bar{u}}_2 = \frac{\lambda}{m} \left( \frac{1}{6} \bar{\bar{p}}_{0z} y^3 + \frac{1}{2} A_z(z, t)y^2 \right) \),

(3.34)
where the no-slip condition \( u_2(y = 0, z, t) = 0 \) has been used. From the normal stress balance (3.7) and tangential stress balance (3.9), we have, to leading order,

\[
\tilde{p}_0 = \frac{J}{\lambda^2} (H + H_{zz})
\]  

(3.35)

and

\[
m\tilde{w}_2 + \tilde{u}_1 + \tilde{w}_{1,zz} = \frac{\beta}{C_\alpha \varepsilon^2} \Gamma_z.
\]  

(3.36)

Consider \( \tilde{w}_2 \)

\[
\tilde{w}_2 \big|_{r = 1 + \varepsilon^2 H} = \frac{a^2 - r^2}{a^2 + m - 1} = \frac{(1 + \varepsilon)^2 - (1 + \varepsilon^2 H)^2}{(1 + \varepsilon)^2 + m - 1} \approx \frac{(2 + \varepsilon)\varepsilon}{m + 2\varepsilon + \varepsilon^2} - \frac{2\varepsilon^2 H}{m + 2\varepsilon + \varepsilon^2}
\]  

(3.37)

where \( \tilde{w}_2 = \frac{(2 + \varepsilon)\varepsilon}{m + 2\varepsilon + \varepsilon^2} \). From the kinematic condition (3.24) and using the Galilean transformation (3.25), we obtain to leading order

\[
\tilde{u}_2 = H_r - \frac{2}{m} HH_z.
\]  

(3.38)

Now, consider \( \tilde{w}_1 \)

\[
\tilde{w}_1 \big|_{r = 1 + \varepsilon^2 H} = 1 - \frac{mr^2}{a^2 + m - 1} = 1 - \frac{(1 + \varepsilon)^2 + m - 1 - m(1 + \varepsilon^2 H)^2}{(1 + \varepsilon)^2 + m - 1} \approx \frac{(2 + \varepsilon)\varepsilon}{m + 2\varepsilon + \varepsilon^2} - \frac{2m\varepsilon^2 H}{m + 2\varepsilon + \varepsilon^2}
\]  

(3.39)

and since \( w_1 = w_2 \) (continuity of velocities) at the interface \( r = 1 + \varepsilon^2 H \), we obtain from (3.39) and (3.37),

\[
-2H + \tilde{w}_1 \big|_{r = 1 + \varepsilon^2 H} = -\frac{2}{m} H.
\]  

(3.40)
Therefore, we conclude from continuity of axial velocities, \( w_1 = w_2 \), and radial velocities, \( u_1 = u_2 \), at the interface, \( r = 1 \) to leading order, that

\[
\tilde{w}_1|_{r=1} = 2H \left( 1 - \frac{1}{m} \right) \quad \text{and} \quad \tilde{u}_1|_{r=1} = 0. \tag{3.41}
\]

We still do not know \( A(z, \tau) \). If we solve the core problem, we can use the normal stress balance (3.7) to find \( A(z, \tau) \). Substituting the core variables into the governing equations (3.1), (3.2), and (3.3), and considering \( Re_1 = \lambda \varepsilon \), we obtain to leading order

\[
\lambda \tilde{p}_{i r} = \nabla^2 \tilde{u}_1 - \frac{\tilde{u}_1}{r^2} \tag{3.42}
\]

\[
\lambda \tilde{p}_{i z} = \nabla^2 \tilde{w}_1 \tag{3.43}
\]

\[
\frac{1}{r} (r \tilde{u}_1)_r + \tilde{w}_1 = 0.
\]

Let us introduce the streamfunction \( \psi \) as

\[
\tilde{u}_1 = -\frac{1}{r} \psi_z \quad \text{and} \quad \tilde{w}_1 = \frac{1}{r} \psi_r. \tag{3.44}
\]

Thus,

\[
\tilde{u}_{1 z} = -\frac{1}{r} \psi_{zz} \tag{3.45}
\]

\[
\tilde{w}_{1 r} = -\frac{1}{r^2} \psi_r + \frac{1}{r} \psi_{rr}. \tag{3.46}
\]

Differentiating (3.33) with respect to \( y \), gives

\[
\tilde{w}_{2 y} = \frac{\lambda}{m} \left( y \tilde{p}_{0 z} + A(z, t) \right). \tag{3.47}
\]

Since \( C_a = \frac{Re_1}{J} \), we have, in this regime, \( C_a \sim \varepsilon \). Considering \( \beta = \varepsilon^3 \beta_0 \) and \( C_a = \varepsilon \tilde{C}_a \), and substituting (3.45), (3.46), (3.47) into the tangential stress balance equation (3.36), yields

\[
\lambda \tilde{p}_{0 z} + \lambda A(z, \tau) = \psi_{zz} + \psi_r - \psi_{rr} + \frac{\beta_0}{C_a} \Gamma_z. \tag{3.48}
\]
The choice $\beta = \varepsilon^3 \beta_0$ is made in order to keep Marangoni effects in the leading order evolution equations. Applying the Fourier transform to the last equation, we obtain the following

$$i\lambda k \hat{p}_0 + \lambda \hat{A} = -k^2 \hat{\psi} + \hat{\psi}_r - \hat{\psi}_{rr} + \frac{ik\beta_0}{C_a} \hat{\Gamma}, \quad (3.49)$$

and solving for $\hat{A}$

$$\hat{A} = -ik \hat{p}_0 - \frac{k^2}{\lambda} \hat{\psi} + \frac{1}{\lambda} \hat{\psi}_r - \frac{1}{\lambda} \hat{\psi}_{rr} + \frac{ik\beta_0}{\lambda C_a} \hat{\Gamma}. \quad (3.50)$$

On the other hand, substituting (3.44) into (3.42) and (3.43), differentiating (3.42) with respect to $z$ and (3.43) with respect to $r$, and eliminating $\hat{p}_1$, we obtain

$$\psi_{rrrr} - \frac{2}{r} \psi_{rrr} + \frac{3}{r^2} \psi_{rr} - \frac{3}{r^3} \psi_r - \frac{2}{r} \psi_{rrr} + 2\psi_{rzz} + \psi_{zzz} = 0. \quad (3.51)$$

Rearranging the last equation, we obtain

$$\frac{\partial^4}{\partial r^4} \psi - \frac{\partial^2}{\partial r^2} \left( \frac{1}{r} \frac{\partial}{\partial r} \right) \psi + 2 \frac{\partial^4}{\partial r^2 \partial z^2} \psi - \frac{1}{r} \frac{\partial^3}{\partial r^3} \psi$$

$$+ \frac{1}{r} \frac{\partial}{\partial r} \left( \frac{1}{r} \frac{\partial}{\partial r} \right) \psi - \frac{2}{r^2} \frac{\partial^3}{\partial r \partial z^2} \psi + \frac{\partial^4}{\partial z^4} \psi = 0, \quad (3.52)$$

which is the creeping flow equation

$$\left( \frac{\partial^2}{\partial r^2} - \frac{1}{r} \frac{\partial}{\partial r} + \frac{\partial^2}{\partial z^2} \right)^2 \psi = 0. \quad (3.53)$$

This is consistent with the fact that $Re_1$ is order $\varepsilon$. The solution of the creeping flow equation (3.53) is most easily accomplished in Fourier space, and is

$$\hat{\psi} = C_1(k)r I_1(kr) + C_2(k)r^2 I_0(kr), \quad (3.54)$$

where $\hat{\psi} = \int_{-\infty}^{\infty} \psi(r,z)e^{-ikz}dz$ is the Fourier transform of $\psi$ and $I_0$, $I_1$ are the modified Bessel functions of order zero and one, respectively. $C_1(k)$ and $C_2(k)$ are two functions
independent from \( r \) and \( z \), to be found. Solution (3.54) excludes contributions from 
\( K_0(kr) \) and \( K_1(kr) \) in order to have finite velocities at \( r = 0 \).

Differentiating (3.54) with respect to \( r \), we have that

\[
\dot{\psi}_r = (C_1k + 2C_2)r I_0(kr) + C_2 kr^2 I_1(kr),
\]

\[
\ddot{\psi}_r = (C_1k + 2C_2 + C_2 k^2 r^2) I_0(kr)
+ (C_1k + 3C_2) kr I_1(kr).
\]

Here, we have used the properties of modified Bessel functions (Abramowitz and Stegun, [1], 1972)

\[
I'_0 = \frac{\partial I_0}{\partial r} = I_1
\]

\[
I'_1 = \frac{\partial I_1}{\partial r} = I_0 - \frac{1}{r k} I_1.
\]

Plugging \( \dot{\psi}_r \) and \( \ddot{\psi}_r \) into (3.50), we find that

\[
\dot{\hat{A}} = -\frac{2k}{\lambda} (kC_1 + C_2) I_1(k) - \frac{2C_2 k^2}{\lambda} I_0(k) + \frac{i k \beta_0}{\lambda C_a} \hat{\Gamma} - i k \dot{\hat{p}}_0
\]

Taking the Fourier transform of (3.34) and plugging (3.57) into it, we have:

\[
\dot{\hat{u}}_2 = -\frac{\lambda k^2 \hat{\beta}_0}{6m} \hat{y}^3 + \frac{i \lambda k}{2m} \hat{A} \hat{y}^2
\]

\[
= \frac{\lambda k^2}{3m} \left( 1 - \frac{1}{3} \hat{y} \right) \hat{y}^2 \hat{p}_0 - \frac{i k^2 (C_1 k + C_2)}{m} \hat{y}^2 I_1(k)
- \frac{i k^3 C_2}{m} \hat{y}^2 I_0(k) - \frac{k^2 \beta_0}{2m C_a} \hat{y}^2 \hat{\Gamma}.
\]

In order to find \( C_1(k) \) and \( C_2(k) \), we consider the Fourier transform of (3.44)

\[
\dot{\psi} = \frac{ir}{k} \int_{-\infty}^{\infty} \hat{u}_1 e^{-i k z} dz \quad \text{and} \quad \dot{\psi}_r = r \int_{-\infty}^{\infty} \hat{\psi}_1 e^{-i k z} dz.
\]

Evaluating \( \dot{\psi} \) and \( \dot{\psi}_r \) at the interface and using (3.41), we obtain, taking the leading order terms,

\[
\dot{\psi}(r = 1) = 0 \quad \text{and} \quad \dot{\psi}_r(r = 1) = 2 \left( 1 - \frac{1}{m} \right) \hat{H}.
\]
Evaluating (3.59) at $r = y = 1$ (the undisturbed interface), we obtain

\[ \frac{\lambda k^2}{3m} \hat{p}_0 - \frac{2ik^2 I_1^2(k)}{m(kI_1^2 - kI_0^2 + 2I_0I_1)} \left( 1 - \frac{1}{m} \right) \hat{H} - \frac{k^2 \beta_0}{2mC_a} \hat{\Gamma} \]  \hspace{1cm} (3.63)

Let us define $d = kI_1^2(k) - kI_0^2(k) + 2I_0(k)I_1(k)$ and $N(k) = \frac{k^2 I_1^2(k)}{d}$. On the other hand, we know that $k^2 \hat{p}_0 = -\hat{p}_{0zz}$ and $k^2 \hat{\Gamma} = -\hat{\Gamma}_{zz}$. Thus,

\[ \hat{u}_2|_{y=1} = -\frac{\lambda}{3m} \hat{p}_{0zz} - \frac{2i}{m} \left( 1 - \frac{1}{m} \right) N(k) \hat{H} + \frac{\beta_0}{2mC_a} \hat{\Gamma}_{zz}. \]  \hspace{1cm} (3.64)

Applying inverse Fourier transform, we obtain

\[ \tilde{u}_2 = -\frac{i}{m\pi} \left( 1 - \frac{1}{m} \right) \int_{-\infty}^{\infty} N(k) \hat{H}(k) e^{ikz} dk \]

\[ - \frac{\lambda}{3m} \hat{p}_{0zz} + \frac{\beta_0}{2mC_a} \hat{\Gamma}_{zz}. \]  \hspace{1cm} (3.65)

Substituting (3.35) and (3.38) into the last equation, we find the interface evolution equation

\[ H_\tau - \frac{2}{m} \frac{\partial}{\partial z} \frac{\partial}{\partial z} H + \frac{i}{m\pi} \left( 1 - \frac{1}{m} \right) \int_{-\infty}^{\infty} N(k) \int_{-\infty}^{\infty} H(z, \tau) e^{ik(z-\bar{z})} d\bar{z} dk \]

\[ + \frac{J}{3m\lambda} \left( H + H_{zz} \right)_{zz} - \frac{\beta_0}{2mC_a} \hat{\Gamma}_{zz} = 0. \]  \hspace{1cm} (3.66)

The last equation without the surfactant diffusion term $-\frac{\beta_0}{2mC_a} \hat{\Gamma}_{zz}$, is exactly the same as equation (28) in (Papageorgiou, Maldarelli, and Rumschitzki [54], 1990). The new term $-\frac{\beta_0}{2mC_a} \hat{\Gamma}_{zz}$ comes from the tangential stress balance (3.9) and is the Marangoni force. It is present along the interface and is directed from regions of high surfactant concentration to regions of low surfactant concentration.
The integral term represents the influence of viscosity stratification, and when \( m = 1 \), that term disappears. Note that the equation (3.66) (without the surfactant diffusion and integral terms) and equation (28) in (Papageorgiou, Maldarelli, and Rumschitzki [54], 1990) (without the integral term) are known as the Kuramoto-Sivashinsky equation (Frenkel, Babchin, Levich, Shlang, and Sivashinsky [19], 1987; Papageorgiou, Maldarelli, and Rumschitzki [54], 1990 and Smyrlis and Papageorgiou [69], 1996).

3.3.2 Derivation of the Concentration of Surfactant Evolution Equation

Consider the non-dimensional concentration of surfactant equation (3.4). Suppose \( Pe \sim O(1/\varepsilon^2) \) and substitute (3.26) and (3.27) into (3.4). Using the fact that \( S(z, t) = 1 + \varepsilon^2 H(z, t) \) and evaluating \( \bar{w}_2 \) at \( r = 1 + \varepsilon^2 H \), we obtain, neglecting high order terms,

\[
\Gamma_t + \frac{\partial}{\partial z} \left[ \bar{w}_2 \Gamma - \frac{2\varepsilon^2 H}{m} \Gamma \right] - \frac{1}{Pe} \Gamma_{zz} = 0.
\]  

(3.67)

Rewriting the second term in the last equation, we obtain

\[
\Gamma_t + \bar{w}_2 \Gamma_z - \frac{2\varepsilon^2}{m} (H\Gamma)_z - \frac{\varepsilon^2}{Pe} \Gamma_{zz} = 0,
\]

(3.68)

where \( \tilde{Pe} = \varepsilon^2 Pe \sim O(1) \). Using the Galilean transformation (3.25) introduced earlier in our analysis of the kinematic condition

\[
\frac{\partial}{\partial t} \rightarrow -\bar{w}_2 \frac{\partial}{\partial z} + \varepsilon^2 \frac{\partial}{\partial r}
\]

(3.69)

we finally obtain, dropping \( \sim \) symbol, the concentration of surfactant evolution equation

\[
\Gamma_z - \frac{2}{m} (H\Gamma)_z - \frac{1}{Pe} \Gamma_{zz} = 0.
\]

(3.70)

This coupled system, (3.66) and (3.70), of evolution equations constitutes an initial value problem for \( H \) and \( \Gamma \), which will be addressed numerically.
In order to perform the numerics, we will first re-scale and study some analytical properties of the system (3.66) and (3.70) in the next section §3.4.

3.4 Re-scaling to Canonical Form and Analytical Properties

3.4.1 Re-scaling to canonical form

We want to go from domains of length $2L$ to domains of length $2\pi$. So, we use the change of variables $x = \frac{L}{\pi} z$ and $\tilde{x} = \frac{L}{\pi} \tilde{z}$. Also, we redefine $H \rightarrow \alpha H$, $\Gamma \rightarrow \gamma \Gamma$, and $\frac{\partial}{\partial t} \rightarrow \beta \frac{\partial}{\partial t}$, where $\alpha$, $\beta$, and $\gamma$ are chosen to make as many coefficients as possible in the nonlinear equations equal to one.

Thus, (3.66) and (3.70) are, using $z$ instead of $x$ and $\tilde{z}$ instead of $\tilde{x}$

\[
\begin{align*}
\alpha \beta H_t & = \alpha^2 \frac{\pi}{L} \frac{2}{m} HH_z \\
& + \frac{i \alpha}{m \pi} \left( \frac{\pi}{L} \right) \left( 1 - \frac{1}{m} \right) \int_{-\infty}^{\infty} N(k) \int_{-\infty}^{\infty} H(z, t) e^{ikz} d\tilde{z} dk \\
& + \left( \frac{\pi}{L} \right)^2 \frac{J \alpha}{3m \lambda} \left( H + \left( \frac{\pi}{L} \right)^2 H_{zz} \right)_{zz} \\
& - \left( \frac{\pi}{L} \right)^2 \gamma \frac{\beta_0}{2m C_a} \Gamma_{zz} = 0. 
\end{align*}
\] (3.71)

\[
\begin{align*}
\beta \gamma \Gamma_t & = \alpha \frac{\pi}{L} \frac{2}{m} (H \Gamma)_z - \left( \frac{\pi}{L} \right)^2 \frac{1}{Pe} \Gamma_{zz} = 0. 
\end{align*}
\] (3.72)

Using the change of variable $\tilde{k} = \frac{L}{\pi} k$, then $dk = \frac{\pi}{L} d\tilde{k}$, (3.71) becomes

\[
\begin{align*}
\alpha \beta H_t & = \alpha^2 \frac{\pi}{L} \frac{2}{m} HH_z \\
& + \frac{i \alpha}{m \pi} \left( 1 - \frac{1}{m} \right) \int_{-\infty}^{\infty} N(\frac{\pi}{L} \tilde{k}) \int_{-\infty}^{\infty} H(z, t) e^{i\tilde{k}(z-\tilde{z})} d\tilde{z} d\tilde{k} \\
& + \left( \frac{\pi}{L} \right)^2 \frac{J \alpha}{3m \lambda} \left( H + \left( \frac{\pi}{L} \right)^2 H_{zz} \right)_{zz} \\
& - \left( \frac{\pi}{L} \right)^2 \gamma \frac{\beta_0}{2m C_a} \Gamma_{zz} = 0. 
\end{align*}
\] (3.73)
Dividing (3.73) by \(-\frac{2\pi\alpha^2}{mL}\), and (3.72) by \(\beta\gamma\), we obtain
\[
- \frac{m\beta}{2\alpha\sqrt{\nu}} H_t + HH_z = \int_{-\infty}^{\infty} N(\sqrt{\nu}k) \int_{-\infty}^{\infty} H(z,t) e^{ik(z-z_d)}dzdk
- \frac{J\sqrt{\nu}}{6\lambda\alpha} (H + \nu H_{zz})_{zz} + \frac{\varepsilon\beta_0\sqrt{\nu}}{4C_\alpha\alpha} \gamma \Gamma_{zz} = 0,
\]

\[
\Gamma_t - 2\frac{\alpha\sqrt{\nu}}{m\beta} (H\Gamma)_z - \frac{\nu}{\beta Pe} \Gamma_{zz} = 0,
\]

where \(\nu = \frac{\pi^2}{L^2}\). Therefore, choosing \(\alpha = -\frac{J\sqrt{\nu}}{6\lambda}\), \(\beta = \frac{J\nu}{3m\lambda}\), and \(\gamma = \frac{C_\alpha J^2\sqrt{\nu}}{9\beta_0\lambda^2}\), we obtain the re-scaled coupled system of the interface and concentration of surfactant evolution equations
\[
H_t + HH_z = \int_{-\infty}^{\infty} N(\sqrt{\nu}k) \int_{-\infty}^{\infty} H(z,t) e^{ik(z-z_d)}dzdk
+ (H + \nu H_{zz})_{zz} = -\Gamma_{zz}
\]

(3.74)

and
\[
\Gamma_t + (H\Gamma)_z = \frac{3m\lambda}{PeJ} \Gamma_{zz}.
\]

(3.75)

Note that this system, in absence of surfactant (\(\Gamma = 0\)) becomes just the equation (28) in (Papageorgiou, Maldarelli, and Rumschitzki [54], 1990), but re-scaled to canonical form. See the first paragraph after equation (3.66) for comments and references that relate (3.74) with the Kuramoto-Sivashinsky equation.

### 3.4.2 Analytical Properties

In this section, we start calculating the canonical form of the perturbed axial velocity \(W_2\). Next, we show that the volume of fluid and the amount of surfactants are conserved. These properties are used to verify the code that we developed in order to do our simulations. We devote the last part of this section to the linear analysis.
of the system (3.74) and (3.75), when \((m = 1)\)

\[
H_r + HH_z + H_{zz} + \nu H_{zzz} = -\Gamma_{zz}
\]

(3.76)

\[
\Gamma_r + (H\Gamma)_z = \eta \Gamma_{zz},
\]

(3.77)

where \(\eta = \frac{3\lambda}{PeJ}\).

**Canonical form of the perturbed axial velocity \(W_2\):** Here, we calculate \(W_2\) which is the canonical form of \(\bar{w}_2\). Evaluating equation (3.33) at \(y = 1\) and considering \(m = 1\), we know that

\[
\bar{w}_2|_{y=1} = \lambda \left( \frac{1}{2} \tilde{p}_0 + A(z, t) \right).
\]

(3.78)

Since \(C_1 = C_2 = 0\) when \(m = 1\), equation (3.57) becomes

\[
\hat{A} = \frac{ik\beta_0}{\lambda C_a} \hat{\Gamma} - i k \hat{p}_0.
\]

(3.79)

Applying the inverse Fourier transform, we obtain

\[
A(z, t) = \frac{\beta_0}{\lambda C_a} \Gamma_z - \tilde{p}_0.
\]

(3.80)

and then (3.78) is

\[
\bar{w}_2|_{y=1} = \lambda \left( -\frac{1}{2} \tilde{p}_0 + \frac{\beta_0}{\lambda C_a} \Gamma_z \right).
\]

(3.81)

Taking the leading order of the equation (3.35) gives us

\[
\tilde{p}_0 = \frac{J}{\lambda^2} (H + H_{zz}),
\]

(3.82)

and substituting (3.82) in (3.81), yields

\[
\bar{w}_2 = -\frac{J}{2\lambda} (H + H_{zz})_z + \frac{\beta_0}{C_a} \Gamma_z.
\]

(3.83)
Re-scaling \( \tilde{w}_2 \) to canonical form \( W_2 \), we use the same re-scaled variables as in §3.4.1 and obtain

\[
W_2 = -\frac{J^2 \nu}{12 \lambda^2} \left[ (H + \nu H_{zz})_z + \frac{4}{3} \Gamma_z \right].
\] (3.84)

**Conserved quantities:** We start considering the system (3.76) and (3.77), then

\[
\frac{d}{d\tau} \left( \int_0^{2\pi} H dz \right) = \int_0^{2\pi} H_t dz
\]

\[
= -\int_0^{2\pi} \left( \Gamma_{zz} + HH_z + H_{zz} + \nu H_{zzzz} \right) dz = 0
\] (3.86)

because of periodicity of \( H \) at the boundaries. Therefore,

\[
\int_0^{2\pi} H dz = \text{constant} = 0
\] (3.87)

if \( H \) has zero mean initially.

\[
\frac{d}{d\tau} \left( \int_0^{2\pi} \Gamma_\tau dz \right) = \int_0^{2\pi} \Gamma_\tau dz
\]

\[
= -\int_0^{2\pi} (\eta \Gamma_{zz} - (HH)_z) dz
\]

\[
= -\Gamma_0 \int_0^{2\pi} H_z dz = 0
\] (3.90)

because \( \Gamma = \Gamma_0 \) is a constant and because of periodicity of \( H \) at the boundaries. Therefore,

\[
\int_0^{2\pi} \Gamma dz = \text{constant} = \Gamma_0 \int_0^{2\pi} dz = 2\pi \Gamma_0.
\] (3.91)

**Linear stability:** The study of the stability of a particular hydrodynamic problem is important in determining its flow type, and the theory of stability is one way to analyze the change from one type of flow to another. Some flows gain stability and others lose it as the parameters of the flow change. In this section, we perform the linear stability of our model considering (3.76) and (3.77). In order to do so, we
consider the undisturbed state

\[ H = 0, \quad \Gamma = \Gamma_0, \quad 0 < \Gamma_0 < 1 \]  

(3.92)

and take normal modes (Drazin and Reid [15], 1999) in the form

\[ H = \hat{H}e^{ikz+\omega t}, \quad \Gamma = \Gamma_0 + \hat{\Gamma}e^{ikz+\omega t}, \]  

(3.93)

where \( k \) is the wave number and \( \omega \) is the growth rate. Substituting (3.93) in (3.76) and (3.77) and retaining only linear terms, we obtain

\[ \omega \hat{H} - k^2 \hat{H} + k^4 \nu \hat{H} = k^2 \hat{\Gamma} \]

(3.94)

\[ \omega \hat{\Gamma} + ik\hat{H}\Gamma_0 = -k^2 \eta \hat{\Gamma}. \]  

(3.95)

Grouping \( \hat{H} \) terms together and \( \hat{\Gamma} \) terms together, yields the system

\[ (\omega - k^2 + \nu k^4)\hat{H} = k^2 \hat{\Gamma} \]

(3.96)

\[ ik\Gamma_0 \hat{H} = - (\omega + \eta k^2) \hat{\Gamma}, \]  

(3.97)

which we solve to obtain

\[ (\omega - k^2 + \nu k^4)(\omega + \eta k^2) = -ik^3 \Gamma_0. \]  

(3.98)

Rewriting the last equation we obtain a quadratic equation for \( \omega \)

\[ \omega^2 + (\eta k^2 - k^2 + \nu k^4)\omega + \eta \nu k^6 - \eta k^4 + ik^3 \Gamma_0 = 0, \]  

(3.99)

which gives two complex values of the growth rate \( \omega_1 \) and \( \omega_2 \)

\[ \omega_{1,2} = -\frac{1}{2}(\eta - 1 + \nu k^2)k^2 \pm \sqrt{\frac{1}{4}(\eta - 1 + \nu k^2)^2 k^4 - \eta k^4(\nu k^2 - 1) - ik^3 \Gamma_0} \]  

(3.100)

Next, we consider \( \Gamma_0 = 1 \) and \( \eta = 1 \) and compute the values of \( \omega_1 \) and \( \omega_2 \) for a range of values of the wave number \( k \). We plot the growth rate \( Re(\omega) \) versus the wave number \( k \), see Figure 3.2(a), (b), and (c).
Considering the growth rate, $Re(\omega_1)$, we can see that it takes positive and negatives values. A cutoff wave number $k_\nu$ exists for each $\nu$ that we studied here.

![Figure 3.2 Plots of the growth rate $Re(\omega)$ versus the wave number $k$ for $\eta = 1$, $\Gamma_0 = 1$. In plot (a) $Re(\omega_1)$ for $\nu = 0.9, 0.2, 0.1, 0.05, 0.01, 0.005, 0.001$, and 0.0007. For (b) $Re(\omega_2)$ for $\nu = 0.9, 0.2, 0.1, 0.05, 0.01, 0.005, 0.001$, and 0.0007. Plot (c) is an amplified plot of $Re(\omega_1)$ and $Re(\omega_2)$ together.](image)

This indicates that for each $\nu$, $Re(\omega_1) < 0$ for $k > k_\nu$. On the other hand, all the values of the growth rate, $Re(\omega_2)$, are negative. Therefore, we can conclude that the solutions are linearly stable for wave numbers $k$ bigger than $k_\nu$. This is consistent with the short wave stabilization supported by the Kuramoto-Sivashinsky equation.
CHAPTER 4
NUMERICAL METHODS

Many numerical methods are based on an infinite expansion of the function to be found, say \( f \), in terms of orthogonal functions \( \phi_k \), so we can express \( f \) as

\[
f = \sum_{k=-\infty}^{\infty} \hat{f}_k \phi_k.
\] (4.1)

We usually approximate periodic functions using Fourier series. In addition, if the function is infinitely smooth and all its derivatives are periodic, the \( k \)-th mode of the Fourier expansion decays faster than any inverse power of \( k \) (algebraic decay).

When there are a sufficient number of Fourier modes to represent all the fundamental structures of the function, then the rapid decay starts; what this means is that after a few modes, the truncated Fourier representation of the function gives a very good approximation of the function. This feature is frequently called the spectral accuracy of the Fourier method.

In this chapter, we develop an implicit scheme to numerically solve the coupled system of equations (3.74) and (3.75) that we derived in chapter 3. The scheme is pseudo-spectral; spectral (Canuto, Hussaini, Quarteroni, and Zang [8], 1988 and Press, Teukolsky, Vetterling, and Flannery [58], 1997) in space and uses a two-step method for the time integrations that achieves second order accuracy.

4.1 Implicit Pseudo-spectral Scheme

The re-scaled coupled system of the interface and concentration of surfactant evolution equations, (3.76) and (3.77) respectively, that we derived in chapter 3 is

\[
H_\tau + HH_z + (H + \nu H_{zz})_{zz} + \Gamma_{zz} = 0
\] (4.2)

\[
\Gamma_\tau + (H\Gamma)_z - \eta \Gamma_{zz} = 0,
\] (4.3)
Applying Fourier transform to the system, we obtain the $k$-th mode system

$$\hat{H}_r^{(k)} + (\nu k^4 - k^2) \hat{H}^{(k)} - k^2 \tilde{\Gamma}^{(k)} = f_1^{(k)}$$  \hspace{1cm} (4.4)$$

$$\hat{\Gamma}_r^{(k)} + \eta k^2 \tilde{\Gamma}^{(k)} = f_2^{(k)},$$  \hspace{1cm} (4.5)

where $f_1^{(k)}$ and $f_2^{(k)}$ are the $k$-th mode of $f_1 = -F(H H_x)$ and $f_2 = -F((H \Gamma)_x)$, respectively, where the operator $F$ denotes Fourier transform. We can express this system in matrix form as follows

$$\begin{bmatrix} \hat{H}^{(k)} \\ \hat{\Gamma}^{(k)} \end{bmatrix}_t + \begin{bmatrix} (\nu k^4 - k^2) & -k^2 \\ 0 & \eta k^2 \end{bmatrix} \begin{bmatrix} \hat{H}^{(k)} \\ \hat{\Gamma}^{(k)} \end{bmatrix} = \begin{bmatrix} f_1^{(k)} \\ f_2^{(k)} \end{bmatrix}.$$  \hspace{1cm} (4.6)

Let us consider the following implicit scheme

$$\begin{bmatrix} \hat{H}^{t+\frac{1}{2}} - \hat{H}^t \\ \hat{\Gamma}^{t+\frac{1}{2}} - \hat{\Gamma}^t \end{bmatrix}^{(k)} = \begin{bmatrix} \hat{H}^{t+\frac{1}{2}} \\ \hat{\Gamma}^{t+\frac{1}{2}} \end{bmatrix}^{(k)} + M(k) \begin{bmatrix} \hat{H}^{t+\frac{1}{2}} + \hat{H}^t \\ \hat{\Gamma}^{t+\frac{1}{2}} + \hat{\Gamma}^t \end{bmatrix}^{(k)}$$  \hspace{1cm} (4.7)

$$\begin{bmatrix} \hat{H}^{t+1} - \hat{H}^t \\ \hat{\Gamma}^{t+1} - \hat{\Gamma}^t \end{bmatrix}^{(k)} = \begin{bmatrix} \hat{H}^{t+1} + \hat{H}^t \\ \hat{\Gamma}^{t+1} + \hat{\Gamma}^t \end{bmatrix}^{(k)} + \frac{1}{2} M(k) \begin{bmatrix} \hat{H}^{t+1} + \hat{H}^t \\ \hat{\Gamma}^{t+1} + \hat{\Gamma}^t \end{bmatrix}^{(k)},$$  \hspace{1cm} (4.8)

where $M(k) = \begin{bmatrix} (\nu k^4 - k^2) & -k^2 \\ 0 & \eta k^2 \end{bmatrix}$. This scheme is spectral in space and half step finite difference in time. The half step is considered to assure that the scheme is $O((\Delta t)^2)$. A proof of this is given in Appendix B. Re-arranging terms with respect to time steps, the system can be written as

$$\begin{bmatrix} I + \frac{\Delta t}{2} M(k) \end{bmatrix} \begin{bmatrix} \hat{H}^{(k)} \\ \hat{\Gamma}^{(k)} \end{bmatrix}^{t+\frac{1}{2}} = \begin{bmatrix} \hat{H}^{(k)} + \frac{\Delta t}{2} f_1^{(k)} \\ \hat{\Gamma}^{(k)} + \frac{\Delta t}{2} f_2^{(k)} \end{bmatrix}^t.$$  \hspace{1cm} (4.9)
\[
\left[ I + \frac{\Delta t}{2} M(k) \right] \begin{bmatrix} \hat{H}^{(k)} \\ \hat{\Gamma}^{(k)} \end{bmatrix}^{t+1} = \left[ I - \frac{\Delta t}{2} M(k) \right] \begin{bmatrix} \hat{H}^{(k)} \\ \hat{\Gamma}^{(k)} \end{bmatrix}^t + \Delta t \begin{bmatrix} f_1^{(k)} \\ f_2^{(k)} \end{bmatrix}^{t+\frac{1}{2}}
\]

In order to solve this system, let us define the matrix \( A \) as

\[
A = I + \frac{\Delta t}{2} M(k) = \begin{bmatrix}
1 + \frac{\Delta t}{2} (\nu k^2 - 1)k^2 & -\frac{\Delta t}{2} k^2 \\
0 & 1 + \frac{\Delta t}{2} \eta k^2
\end{bmatrix}.
\]  

(4.11)

Then,

\[
A^{-1} = \frac{1}{|A|} \begin{bmatrix}
1 + \frac{\Delta t}{2} \eta k^2 & \frac{\Delta t}{2} k^2 \\
0 & 1 + \frac{\Delta t}{2} (\nu k^2 - 1)k^2
\end{bmatrix}.
\]  

(4.12)

In order to simplify our notation, we introduce the following two symbols \( \varrho = \frac{\Delta t}{2} (\nu k^2 - 1)k^2 \) and \( \varphi = \frac{\Delta t}{2} \eta k^2 \). So,

\[
A^{-1} \left[ I - \frac{\Delta t}{2} M(k) \right] = \frac{1}{|A|} \begin{bmatrix}
(1 + \varphi)(1 - \vartheta) & \Delta tk^2 \\
0 & (1 + \vartheta)(1 - \varphi)
\end{bmatrix},
\]  

(4.13)

where \( |A| = (1 + \vartheta)(1 + \varphi) \). In this way our system becomes

\[
\begin{align*}
\begin{bmatrix} \hat{H}^{(k)} \\ \hat{\Gamma}^{(k)} \end{bmatrix}^{t+\frac{1}{2}} &= A^{-1} \begin{bmatrix} \hat{H}^{(k)} + \frac{\Delta t}{2} f_1^{(k)} \\ \hat{\Gamma}^{(k)} + \frac{\Delta t}{2} f_2^{(k)} \end{bmatrix}^t \\
\begin{bmatrix} \hat{H}^{(k)} \\ \hat{\Gamma}^{(k)} \end{bmatrix}^{t+1} &= A^{-1} \left[ I - \frac{\Delta t}{2} M(k) \right] \begin{bmatrix} \hat{H}^{(k)} \\ \hat{\Gamma}^{(k)} \end{bmatrix}^t + A^{-1} \Delta t \begin{bmatrix} f_1^{(k)} \\ f_2^{(k)} \end{bmatrix}^{t+\frac{1}{2}}
\end{align*}
\]

(4.14)

(4.15)

Expressing the system in single equations, we obtain

\[
\begin{align*}
\hat{H}^{(k)}_{t+\frac{1}{2}} &= \frac{1}{1 + \varphi} \left[ \left( \hat{H}^{(k)} + \frac{\Delta t}{2} f_1 \right)^t + \frac{k^2 \Delta t}{2(1 + \varphi)} \left( \hat{\Gamma}^{(k)} + \frac{\Delta t}{2} f_2 \right)^t \right] \\
\hat{\Gamma}^{(k)}_{t+\frac{1}{2}} &= \frac{1}{1 + \vartheta} \left( \hat{\Gamma}^{(k)} + \frac{\Delta t}{2} f_2 \right)^t
\end{align*}
\]

(4.16)
and the second step system

\[
\begin{align*}
\hat{H}^{(k)+1} &= \frac{1}{1+\varphi} \left((1-\varphi)\hat{H}^{(k)} + \frac{k^2\Delta t}{(1+\varphi)}\hat{H}^{(k)+\frac{1}{2}} + \frac{(\Delta t)^2 k^2}{2(1+\varphi)}\hat{f}_2^{(k)+\frac{1}{2}}\right) \\
\hat{\Gamma}^{(k)+1} &= \frac{1}{1+\varphi} \left((1-\varphi)\hat{\Gamma}^{(k)} + \Delta t\hat{f}_2^{(k)+\frac{1}{2}}\right).
\end{align*}
\]

(4.17)

In the next section, we present an algorithm of our scheme.

### 4.2 Algorithm: Implicit Pseudo-spectral using Finite Differences

In this section, we present an algorithm for the implicit pseudo-spectral scheme that we developed in §4.1 to solve our problem.

**Algorithm:**

To approximate the solution of

\[
H_t + HH_z + (H + \nu H_{zz})_{zz} + \Gamma_{zz} = 0
\]

\[
\Gamma_t + (H\Gamma)_z - \eta \Gamma_{zz} = 0,
\]

where \(\nu = \frac{\pi^2}{L^2}\) and \(\eta = \frac{3\lambda}{PeJ}\).

**INPUT:** integer \(n\), \(finalt\) (final time), \(\Delta t\), \(\nu\), and \(\eta\).

**OUTPUT:** \(H\) and \(\Gamma\)

**Step 1** Set \(\Delta z = 2\pi/n\)

\[t = 0\]

**Step 2** Compute: Grid points \(z_j = j\Delta z, \quad j = 0, \ldots, n - 1\)

Initial Conditions \(H_j^0 = -\sin(z_j)\)

and \(\Gamma_j^0 = \Gamma_0\) (constant) \(0 < \Gamma_0 \leq 1\)

**Step 3** While \(t < finalt\) do Steps 4-14
(First system step \( t + \frac{1}{2} \))

Step 4 Compute \( H \Gamma \) in physical space.

Step 5 Compute \( H_z \) and \( (H \Gamma)_z \) in Fourier space.

Step 6 Compute \( HH_z \) in physical space.

Step 7 Transform \( HH_z \) in Fourier space.

Step 8 For each \( k \)-th mode compute:
\[
\varrho = \frac{\Delta t}{2} (\nu k^2 - 1) k^2 \quad \text{and} \quad \varphi = \frac{\Delta t}{2} \eta k^2
\]

\[
\hat{\Gamma}^{(k)^{t+\frac{1}{2}}} = \frac{1}{1 + \varphi} \left( \hat{\Gamma}^{(k)} + \frac{\Delta t}{2} f_2 \right)^t
\]

\[
\hat{H}^{(k)^{t+\frac{1}{2}}} = \frac{1}{1 + \varphi} \left[ \left( \hat{H}^{(k)} + \frac{\Delta t}{2} f_1 \right)^t + \frac{\Delta t k^2}{2(1 + \varphi)} \left( \hat{\Gamma}^{(k)} + \frac{\Delta t}{2} f_2 \right)^t \right]
\]

(Second system step \( t + 1 \))

Step 9 Compute \( H \Gamma \) in physical space.

Step 10 Compute \( H_z \) and \( (H \Gamma)_z \) in Fourier space.

Step 11 Compute \( HH_z \) in physical space.

Step 12 Transform \( HH_z \) in Fourier space.

Step 13 For each \( k \)-th mode compute:
\[
\varrho = \frac{\Delta t}{2} (\nu k^2 - 1) k^2 \quad \text{and} \quad \varphi = \frac{\Delta t}{2} \eta k^2
\]

\[
\hat{\Gamma}^{(k)^{t+1}} = \frac{1}{1 + \varphi} \left[ (1 - \varrho) \hat{\Gamma}^{(k)^t} + \Delta t f_2^{(k)^{t+\frac{1}{2}}} \right]
\]

\[
\hat{H}^{(k)^{t+1}} = \frac{1}{1 + \varrho} \left[ (1 - \varrho) \hat{H}^{(k)^t} + \frac{\Delta t k^2}{(1 + \varphi)} \hat{\Gamma}^{(k)^t} + \Delta t f_1^{(k)^{t+\frac{1}{2}}} + \frac{(\Delta t)^2 k^2}{2(1 + \varphi)} f_2^{(k)^{t+\frac{1}{2}}} \right]
\]

Step 14 Update time: \( t = t + \Delta t \)

Step 15 Output \( H^t, \Gamma^t \).

Step 16 End.
4.3 Numerical Experiments

In this section, we explain how we implemented the algorithm that we developed in §4.2 and carried out the numerical simulations. We coded in the FORTRAN programming language (Nyhoff and Leestma [50], 1992) and also used a package of FORTRAN subroutines that perform the Fast Fourier Transform of periodic boundaries, developed by Swarztrauber ([73], 1978).

We have performed extensive numerical experiments, examining over 200 values of the parameter $\nu = \frac{\pi^2}{L^2}$ ranging from $\nu = 1.0$ to $\nu = 0.000705$. In order to implement Fast Fourier Transform (FFT) routines, the number of nodes, $n$, must be a power of 2 and in our case, $n$ ranged from $n = 64$ to $n = 2048$ increasing the number of operations in the computation as $n$ is increased.

In order to verify our code, we considered the case $\Gamma = 0$ and were able to reproduce the results of Papageorgiou and Smyrlis ([56], 1991). We also checked that the conserved quantities, which we discussed in §3.4.2,

$$
\int_0^{2\pi} H dz = 0 \quad \text{and} \quad \int_0^{2\pi} \Gamma dz = 2\pi \Gamma_0,
$$

are satisfied.

For many cases, long computations are required in order to determine the nature of the attractor (see Appendix C for a definition of attractor) and accurately classify it. This happens especially near the boundaries between intervals of $\nu$, where the attractor changes from, let us say, a traveling wave attractor to a time-periodic attractor, or vice versa.

From $\nu = 1.0$ to $\nu = 0.098$, we used as initial conditions $H_0 = -\sin z$ and $\Gamma_0 = 1$. For $\nu = 0.097$, we obtained numerical instabilities even for different amplitudes of $H_0$, different values of $\Gamma_0$, different values of $n$, the number of nodes in space, and different values of the time step. So, we decided to use continuation which means that we use the results of $H$ for a current value of the parameter $\nu$ as the initial condition.
for the next value of $\nu$ and let the code run. Using this technique, we were able to
overcome the problem of numerical instability of the solutions because now the initial
condition $H_0$ is sufficiently near the attractor of the new value of $\nu$.

For example, choosing the solution for $\nu = 0.1$ as initial condition for the next
value of $\nu = 0.099$, and then using the solution for $\nu = 0.099$ as initial condition for
the next value of $\nu = 0.098$ and so on, we were able to find solutions for all values, in
parameter space, of $\nu$ from 0.099 to 0.000705.

4.4 Characterization of Attractors

In order to classify an attractor (see Appendix C for a definition of attractor), we
carried out several hundreds of numerical experiments and analyzed the time evolution
of the interface and concentration of surfactant profiles, their energies ($L^2$-norm), the
phase-plane of the energies, the maxima/minima of the energies, and the return map
of the maxima/minima of the energies.

In Appendix C, we provide a full description of our diagnostics and the tools we
use in the characterization of attractors. The time evolution of the profile energy ($L^2$-
norm) is illustrative of the nature of various attractors. Stationary and traveling wave
attractors evolve to a constant value of energy in time. Time-periodic, quasi-periodic,
and chaotic attractors evolve to an oscillatory state of energy in time.

The interface profile and the concentration of surfactant profile are helpful to
classify an attractor as stationary or steady-state traveling wave by comparing the
profiles for two different times. In the case of a space-periodic traveling wave attractor
we can compute the wave speed using the profiles for those two different times, once
transients have died out and the solution reaches its long time behavior.

In order to classify time-periodic attractors (see Appendix C), we analyzed
the energy phase-plane, the return map of the energy maxima and minima, and the
integral of the energy $I_E$ defined by

$$I_E = \frac{1}{\tau} \int_{t_0}^{t_0+\tau} E(t) dt,$$

(4.19)

where $t_0$ is the time at the beginning of the period, $\tau$ is the period of the wave, and $E(t)$ is the energy.

In general, in our problem, we obtained an alternating behavior between a constant energy in time (space-periodic traveling wave attractors) from one interval to an oscillatory energy in time (time-periodic traveling attractors) in the next interval, and so on.

In the cases of space-periodic traveling waves, we have found that for the first interval $0.2 < \nu$ the space-periodic traveling waves were **fully-modal**, i.e. $2\pi$-periodic in space. For the second interval with traveling wave behavior, $0.055 < \nu < 0.099$, we obtained **bi-modal** traveling waves, i.e. $\pi$-periodic in space. In the third interval with space-periodic traveling wave behavior, $0.014 < \nu < 0.04$, the attractors were **tetra-modal**; i.e. $\pi/2$-periodic in space traveling wave attractors. For the next interval of attractors with space-periodic traveling wave behavior, $0.00375 < \nu < 0.01$, we obtained, as we expected, **octa-modal** traveling waves, i.e. $\pi/4$-periodic traveling waves attractors.

We checked and found that, when $\nu$ is small enough, there is a relationship that we find in (Papageorgiou, Papanicolaou, and Smyrlis [55], 1992 and Smyrlis and Papageorgiou [69], 1996) which gives us an idea of the behavior, wave speed, and amplitude of the traveling wave attractor in a specific interval. In what follows, we show that the following family of solutions can be constructed from a given traveling wave $U(x - ct; \nu)$,

$$u(x, t) = \rho U(\rho(x - ct); \rho^2 \nu) + c,$$

(4.20)
where $U$ is the steady state wave that we know with its parameters $\nu$, and $c$; and $\rho$ is an integer number for which we find a new steady attractor $u$; this new attractor $u$ corresponds to $\nu_\rho = \nu/\rho^2$, has period $2\pi/\rho$, and amplitude increased by $\rho$. This property is used to generate steady states of the Kuramoto-Sivashinsky equation at geometrically decreasing values of $\nu$; for details see Papageorgiou, Papanicolaou, and Smyrlis ([55], 1992) and Smyrlis and Papageorgiou ([69], 1996). In our problem we use (4.20) to find a good approximation to the initial value of the profile to be used to find the solution corresponding to the new value of $\nu_\rho = \nu/\rho^2$. See Appendix C for more details.

In this way, we were able to predict the next interval, $0.0009 < \nu < 0.0025$, where the attractors should be decahexa-modal, i.e. $\pi/8$-periodic in space. We were able to check numerically the theoretical predictions (see Table 4.4).

<table>
<thead>
<tr>
<th>Interval of $\nu$</th>
<th>Behavior</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(0, 0.200]$</td>
<td>Fully modal traveling</td>
</tr>
<tr>
<td>$[0.099, 0.190]$</td>
<td>Time-periodic traveling</td>
</tr>
<tr>
<td>$[0.055, 0.099]$</td>
<td>Bi-modal traveling</td>
</tr>
<tr>
<td>$[0.041, 0.054]$</td>
<td>Time-periodic traveling</td>
</tr>
<tr>
<td>$[0.014, 0.040]$</td>
<td>Tetra-modal traveling</td>
</tr>
<tr>
<td>$[0.011, 0.013]$</td>
<td>Time-periodic traveling</td>
</tr>
<tr>
<td>$[0.00375, 0.01]$</td>
<td>Octa-modal traveling</td>
</tr>
<tr>
<td>$[0.00275, 0.0035]$</td>
<td>Time-periodic traveling</td>
</tr>
<tr>
<td>$[0.00090, 0.0025]$</td>
<td>Decahexa-modal traveling</td>
</tr>
<tr>
<td>$[0.000705, 0.0008]$</td>
<td>Time-periodic traveling</td>
</tr>
</tbody>
</table>

For the case of time-periodic traveling waves, found in the first interval $0.099 \leq \nu \leq 0.19$, the number of energy maxima and minima of the solutions goes from one to
three and then from three to five, as $\nu$ decreases. In the second interval, $0.041 \leq \nu \leq 0.054$, of time-periodic traveling waves, the number of energy maxima and minima of the solutions goes from one to two when $\nu$ goes from 0.049 to 0.04875. For the third window of time-periodic traveling waves, $0.011 \leq \nu \leq 0.013$, the solutions have only two energy maxima and two energy minima for all the values of $\nu$ in this interval. In the fourth interval of time-periodic traveling waves, $0.00275 \leq \nu \leq 0.0035$, we have only one case of one maximum and one minimum, which is for $\nu = 0.0035$, and for the other values of $\nu$, we have two energy maxima and minima. In the last window, $0.000705 \leq \nu \leq 0.0008$, of time-periodic traveling waves, the solutions have only two energy maxima and two energy minima for all values of $\nu$ in the interval.
CHAPTER 5

NUMERICAL RESULTS

5.1 Fully Modal Traveling Attractors: $0.2 \leq \nu$

For this window, solutions get attracted to $2\pi$-periodic traveling waves. In our simulations, we use $\eta = 1$, $\beta = 1$, $\Gamma_0 = 1$ and $H_0 = -\beta \sin z$. We also use $\Delta t = 10^{-3}$, $time = 100$, and 64 nodes in space for $0.7 \leq \nu$ and 128 nodes in space for $0.2 \leq \nu \leq 0.6$. The interface “energy”, $E_H(t) = \int_0^{2\pi} H^2(z, t) dz$, is defined as in §4.4.

In order to analyze the solutions, we classify them in two categories. First, those solutions that travel forward and have positive wave speed ($c$), and another for those that travel backwards, with wave speed ($c$) negative. Table 5.1 gives an overview of such solutions. It includes several values of $\nu$ and provides the corresponding wave speed $c$, energy $E_H$, and number of nodes $n$.

In the first category, $0.5 \leq \nu \leq 1.0$, the wave speed decreases monotonically from approximately $c = 0.540$ for $\nu = 1.0$ to $c = 0.114$ for $\nu = 0.5$. The wave is traveling forward and the energy, $E_H$, is increasing slowly, i.e. with a decreasing rate of growth. In the second category, $0.2 \leq \nu \leq 0.4$, the wave speed decreases from approximately $c = -0.153$ for $\nu = 0.4$ to $c = -1.270$ for $\nu = 0.2$. The wave is moving backward ($c$ is negative) and the energy, $E_H$, growing with an increasing rate of growth.

In Figure 5.1, we plot the traveling wave profiles of the interface ($H$), the concentration of surfactant ($\Gamma$) and the corresponding axial velocity ($W_2$) for (a) $\nu = 0.9$ and (b) $\nu = 0.3$. The quantity $W_2$ is the perturbation axial velocity that arises to leading order from our asymptotic solution and which can be written completely in terms of $H$ and $\Gamma$ (see §3.4.2) as

$$W_2 = (\nu H_{zz} + H)_z + \frac{4}{3} \Gamma_z.$$
Table 5.1  Wave speed (c), interface energy ($E_H$), and number of nodes (n) for the fully modal traveling case, $0.2 \leq \nu$. Here, $\eta = 1$, $\beta = 1$, $\Gamma_0 = 1$, $H_0 = -\beta \sin z$, $\Delta t = 10^{-3}$, $n = 64, 128$, and $\text{time} = 100$.

<table>
<thead>
<tr>
<th>$\nu$</th>
<th>1.0</th>
<th>0.9</th>
<th>0.8</th>
<th>0.7</th>
<th>0.6</th>
<th>0.5</th>
<th>0.4</th>
<th>0.3</th>
<th>0.2</th>
</tr>
</thead>
<tbody>
<tr>
<td>$c$</td>
<td>0.540</td>
<td>0.475</td>
<td>0.400</td>
<td>0.319</td>
<td>0.234</td>
<td>0.114</td>
<td>-0.153</td>
<td>-0.635</td>
<td>-1.270</td>
</tr>
<tr>
<td>$E_H$</td>
<td>15.22</td>
<td>19.75</td>
<td>24.44</td>
<td>28.45</td>
<td>30.95</td>
<td>32.24</td>
<td>36.86</td>
<td>52.35</td>
<td>71.21</td>
</tr>
<tr>
<td>$n$</td>
<td>64</td>
<td>64</td>
<td>64</td>
<td>64</td>
<td>128</td>
<td>128</td>
<td>128</td>
<td>128</td>
<td>128</td>
</tr>
</tbody>
</table>

The relative positioning of the interfacial and surfactant concentration profiles can be explained physically by consideration of the perturbation velocity $W_2$. The mechanism described next is at play for the multi-modal traveling wave profiles found at decreasing values of $\nu$.

The perturbation velocity $W_2$ is solely responsible, to leading order, for the non-uniform convection of surfactants along the interface and creation of Marangoni forces, since we are in a traveling frame of reference. Inspection of Figure 5.1 (but see also Figures 5.4, 5.6, 5.8, and 5.10 for multi-modal waves) indicates that there is a stagnation point where $W_2 = 0$ just ahead of the interfacial wave maximum. The local flow is positive to the left of this point and negative to its immediate right, and so causes surfactant to be swept to its vicinity. This in turn creates a maximum in surfactant concentration just ahead of the interfacial wave, and this is clearly seen from the Figures. Consequently, the position of the peak in surfactant concentration (and hence the distribution of Marangoni forces) is slaved to the position of the interfacial maximum. Inspection of Figures 5.1(a) and (b) (and indeed parts (a) and (b) of Figures 5.4, 5.6, 5.8, and 5.10 also), and noting that the direction of travel of the waves in parts (b) are negative, leads to the conclusion that for left traveling waves the position of surfactant peaks is behind the wave maximum as opposed to ahead of it for right traveling waves. We should note, however, that this change is not
of significant physical importance since our system of equations is derived in a frame
of reference which is traveling with speed of order \( \varepsilon \) (the base-flow velocity at the
undisturbed film position) whereas the scaled computed speeds provide corrections
of order \( \varepsilon^2 \). In the physical frame of reference, then, all waves are right-moving and
the peaks in surfactant concentration are to be found ahead of the interfacial wave
maximum.

\[\text{Figure 5.1} \quad \text{Profiles of interface } (H), \text{ concentration of surfactant } (\Gamma), \text{ and axial}
velocity \( (W_2) \) for } \eta = 1, \beta = 1, \Gamma_0 = 1, H_0 = -\beta \sin z, \Delta t = 10^{-3}, \text{ and time=100.}
For (a) \( \nu = 0.9, n = 64, \) and \( c = 0.4750 \). For (b) \( \nu = 0.3, n = 128, \) and \( c = -0.6350 \).

5.2 Time-Periodic Attractors: \( 0.099 \leq \nu \leq 0.19 \)

At large times, the solutions of our system for values of \( \nu \) in this interval are attracted
to time-periodic traveling orbits. In our runs we use 128 nodes in space, \( \Delta t = 10^{-3}, \)
\( \text{time} = 1000, \eta = 1, \beta = 1, \Gamma_0 = 1, \) and \( H_0 = -\beta \sin z \). For our analysis of
the behavior of the solutions, in this window we classify them in three categories.
In Appendix C we give a description of the diagnostics of the solutions. The first
category in our classification is for solutions that have only one value for maxima
and only one value for minima in the energy \( (E_H) \) profile. The second category for
solutions that have three values for maxima and three values for minima in the energy
The third category in this window is for solutions that have five values for maxima and five values for minima in the energy ($E_H$) profile, see Table 5.2.

**Table 5.2** Period ($\tau$), integral of energy ($I_E$), and number of maxima and minima for $0.099 \leq \nu \leq 0.19$.

<table>
<thead>
<tr>
<th>$\nu$</th>
<th>$\tau$</th>
<th>Integral of Energy $I_E$</th>
<th>Max./min.</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.190</td>
<td>1.380</td>
<td>11.594</td>
<td>1</td>
</tr>
<tr>
<td>0.180</td>
<td>1.345</td>
<td>11.713</td>
<td>1</td>
</tr>
<tr>
<td>0.170</td>
<td>1.295</td>
<td>11.607</td>
<td>1</td>
</tr>
<tr>
<td>0.160</td>
<td>1.251</td>
<td>11.583</td>
<td>1</td>
</tr>
<tr>
<td>0.150</td>
<td>1.218</td>
<td>11.656</td>
<td>1</td>
</tr>
<tr>
<td>0.140</td>
<td>1.190</td>
<td>11.791</td>
<td>1</td>
</tr>
<tr>
<td>0.130</td>
<td>1.159</td>
<td>11.890</td>
<td>1</td>
</tr>
<tr>
<td>0.120</td>
<td>1.117</td>
<td>11.807</td>
<td>1</td>
</tr>
<tr>
<td>0.118</td>
<td>1.077</td>
<td>11.761</td>
<td>1</td>
</tr>
<tr>
<td>0.115</td>
<td>1.443</td>
<td>18.085</td>
<td>3</td>
</tr>
<tr>
<td>0.114</td>
<td>1.482</td>
<td>19.079</td>
<td>3</td>
</tr>
<tr>
<td>0.112</td>
<td>1.518</td>
<td>20.273</td>
<td>3</td>
</tr>
<tr>
<td>0.111</td>
<td>3.004</td>
<td>56.396</td>
<td>5</td>
</tr>
<tr>
<td>0.110</td>
<td>2.949</td>
<td>55.839</td>
<td>5</td>
</tr>
<tr>
<td>0.100</td>
<td>3.053</td>
<td>64.037</td>
<td>5</td>
</tr>
<tr>
<td>0.099</td>
<td>3.031</td>
<td>64.126</td>
<td>5</td>
</tr>
</tbody>
</table>

Periodicity is determined by examination of the energy phase-plane ($E$ versus $dE/dt$), and the period, $\tau$, is determined from the maxima and minima of energy versus time, see Appendix C. The bifurcations from one maximum and one minimum to three maxima and three minima to five maxima and five minima states are not
smooth (a smooth bifurcation would result in the case of a period doubling, for example). Instead, what is happening is that one periodic state becomes unstable to a different coexisting periodic state. One way to quantify this is to examine the integral $I_E = \frac{1}{\tau} \int_{t_0}^{t_0+\tau} E(t) dt$, where $\tau$ is the period of oscillation and $t_0$ is an arbitrary time chosen so that the transients have died out. Values of $I_E(\tau, \nu)$ are given in Table 5.2 and it is clear from these values that the 1, 3, and 5 maxima/minima solution branches are not smoothly connected. We note that our numerical methods are based on initial value problems and cannot follow the unstable periodic branches.

More specifically, looking at Table 5.2 we can observe that the energy of the solutions in the first category, sub-interval $0.116 \leq \nu \leq 0.19$, has only one maximum and one minimum. Figure 5.2(a) is a plot of the maxima and minima for $\nu = 0.116$. The period starts at approximately a value of $\tau = 1.38$ for $\nu = 0.190$ and decreases slowly to approximately a value of 1.097 for $\nu = 0.116$. All the values of $I_E$ in this sub-interval change smoothly indicating that the solutions in this sub-interval remain in the same attractor.

In the second category, which is the next sub-interval of $\nu$ in parameter space, $0.112 \leq \nu \leq 0.115$, the energy attains three maxima and three minima, see Figure 5.2(b). Since it is not easy to see the three maxima (color blue in Figure 5.2(b)), we plot the return map for the maxima and minima, Figure 5.2(d). The period increases monotonically from $\tau = 1.443$ for $\nu = 0.115$ to $\tau = 1.5177$ for $\nu = 0.112$. We observe that there is a jump in period from $\tau = 1.097$ for $\nu = 0.116$ (first category) to $\tau = 1.443$ for $\nu = 0.115$ (second category), see Table 5.2. Also, we can notice that there is a drastic change in $I_E$ between $\nu = 0.116$ and $\nu = 0.115$; this indicates that the solutions in this second sub-interval are drawn by another attractor.

Going to the next category, the next value of $\nu$, in parameter space, is $\nu = 0.111$. It seems that a period doubling occurs, but we shall see that this is not the case. Considering the sub-interval, $0.099 \leq \nu \leq 0.111$, where the energy changes from
Figure 5.2  Plots of maxima and minima of $E_H$ versus time for $\eta = 1$, $\beta = 1$, $\Gamma_0 = 1$, $H_0 = -\beta \sin z$, $n = 128$, $\Delta t = 10^{-3}$, and time $= 1000$. In plot (a) $\nu = 0.116$, the solution has one maximum and one minimum. For (b) $\nu = 0.112$, with three maxima and three minima. In (c) $\nu = 0.111$, solution has five maxima and five minima. In (d) we plot the return map of the maxima and minima for the case (b) to make sure that case (b) really has three maxima.
three to five maxima and five minima (see Figure 5.2(c) for a plot of the maxima and
minima of $\nu = 0.111$) at $\nu = 0.111$, the period is $\tau = 3.004$ and keeps roughly the
same value throughout the sub-interval (see Table 5.2) having a value of $\tau = 3.031$
for $\nu = 0.099$. This sub-interval starts with a period which is almost double that
corresponding to $\nu = 0.112$ from the second category.

Looking at the energy phase-planes, Figure 5.3 for $\nu = 0.116$, $\nu = 0.112$, and
$\nu = 0.111$, and examining the values of $I_E$ in Table 5.2 we conclude that the solutions
in this sub-interval are approaching a different attractor from the attractors of the
two first sub-intervals. Therefore, there is no period doubling taking place.

From the value of $\nu = 0.099$ on, we use continuation in $H$ and $\Gamma$; i.e., we take
the final solution for $H$, corresponding to the last computed $\nu$, and use it as initial
condition to find the solutions $H$ and $\Gamma$ for the next value of $\nu$. In this way, we
compute solutions for smaller values of $\nu$, and avoid stability problems which can
occur in the transient dynamics.

5.3 Bi-Modal Traveling Attractors: $0.055 \leq \nu \leq 0.099$
In this interval, the solutions are attracted to $\pi$-periodic traveling waves or bi-modal
traveling waves. In our simulations, we use 128 nodes in space, the values of $\eta = 1, \Gamma_0 = 1, H_0$ in continuation format, $\Delta t = 10^{-3}$, and $time = 100$. The wave
speed decreases monotonically, and is negative which means that the wave is moving
backward, from approximately $-0.170$ for $\nu = 0.099$ to $-2.095$ for $\nu = 0.055$, see
Table 5.3 for details. The energy, $E_H$, defined in §5.1, increases as $\nu$ decreases.

In Figure 5.4, we plot the profiles of the interface ($H$), concentration of
surfactant ($\Gamma$), and axial velocity ($W_2$) for (a) $\nu = 0.095$ and (b) $\nu = 0.058$. For
a physical explanation of the relative positions of the three plotted profiles, see §5.1.
Figure 5.3  Phase-plans of $E_H$ for $\eta = 1$, $\beta = 1$, $\Gamma_0 = 1$, $H_0 = -\beta \sin z$, $n = 128$, $\Delta t = 10^{-3}$, and time = 1000. In plot (a) $\nu = 0.116$ and the period is $\tau = 1.097$. For (b) $\nu = 0.112$ with $\tau = 1.518$. In (c) $\nu = 0.111$ and the period is $\tau = 3.004$. We start plotting $(E_H, dE_H/dt)$ from $t = 500$. 
Table 5.3 Wave speed \((c)\) and interface energy \((E_H)\) for \(0.055 \leq \nu \leq 0.099\). Bimodal traveling case. Here, \(\eta = 1, \Gamma_0 = 1, H_0\) in continuation format, \(\Delta t = 10^{-3}\), \(n = 128\), and \(\text{time} = 100\).

<table>
<thead>
<tr>
<th>(\nu)</th>
<th>0.099</th>
<th>0.095</th>
<th>0.090</th>
<th>0.085</th>
<th>0.080</th>
</tr>
</thead>
<tbody>
<tr>
<td>(c)</td>
<td>-0.170</td>
<td>-0.348</td>
<td>-0.420</td>
<td>-0.630</td>
<td>-0.883</td>
</tr>
<tr>
<td>(E_H)</td>
<td>110.503</td>
<td>110.937</td>
<td>114.412</td>
<td>122.672</td>
<td>136.262</td>
</tr>
<tr>
<td>(\nu)</td>
<td>0.075</td>
<td>0.070</td>
<td>0.065</td>
<td>0.060</td>
<td>0.055</td>
</tr>
<tr>
<td>(c)</td>
<td>-1.141</td>
<td>-1.411</td>
<td>-1.664</td>
<td>-1.878</td>
<td>-2.095</td>
</tr>
<tr>
<td>(E_H)</td>
<td>153.719</td>
<td>172.426</td>
<td>189.886</td>
<td>204.670</td>
<td>216.389</td>
</tr>
</tbody>
</table>

Figure 5.4 Profiles of interface \((H)\), concentration of surfactant \((\Gamma)\), and axial velocity \((W_2)\) in the bimodal traveling case. \(\eta = 1, \Gamma_0 = 1, H_0\) in continuation format, \(\Delta t = 10^{-3}\), \(n = 128\), and \(\text{time} = 100\). For (a) \(\nu = 0.095, c = -0.3480\). For (b) \(\nu = 0.058, c = -1.9640\).
5.4 Time-Periodic Attractors: $0.041 \leq \nu \leq 0.054$

In this window, time periodic traveling attractors are found. In our runs, we use $\eta = 1$, $\Gamma_0 = 1$ and $H_0$ in continuation format. We also use 128 nodes in space, $\Delta t = 10^{-3}$, and time = 1000. For the analysis of the behavior of the solutions, in this window we classify them in two categories. The first category corresponds to solutions that have one value for maxima and one value for minima in the energy ($E_H$) profile. The second category corresponds to solutions that have two values for maxima and two values for minima in the energy ($E_H$) profile, see Table 5.4.

<table>
<thead>
<tr>
<th>$\nu$</th>
<th>$\tau$</th>
<th>Integral of Energy ($I_E$)</th>
<th>Max./min.</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.054</td>
<td>0.413</td>
<td>206.428</td>
<td>1</td>
</tr>
<tr>
<td>0.053</td>
<td>0.443</td>
<td>211.179</td>
<td>1</td>
</tr>
<tr>
<td>0.052</td>
<td>0.462</td>
<td>212.291</td>
<td>1</td>
</tr>
<tr>
<td>0.051</td>
<td>0.474</td>
<td>219.972</td>
<td>1</td>
</tr>
<tr>
<td>0.050</td>
<td>0.481</td>
<td>221.783</td>
<td>1</td>
</tr>
<tr>
<td>0.049</td>
<td>0.484</td>
<td>228.833</td>
<td>1</td>
</tr>
<tr>
<td>0.048</td>
<td>0.485</td>
<td>213.535</td>
<td>2</td>
</tr>
<tr>
<td>0.047</td>
<td>0.483</td>
<td>216.515</td>
<td>2</td>
</tr>
<tr>
<td>0.046</td>
<td>0.479</td>
<td>231.607</td>
<td>2</td>
</tr>
<tr>
<td>0.045</td>
<td>0.474</td>
<td>236.928</td>
<td>2</td>
</tr>
<tr>
<td>0.044</td>
<td>0.466</td>
<td>225.836</td>
<td>2</td>
</tr>
<tr>
<td>0.043</td>
<td>0.456</td>
<td>240.260</td>
<td>2</td>
</tr>
<tr>
<td>0.042</td>
<td>0.444</td>
<td>242.422</td>
<td>2</td>
</tr>
<tr>
<td>0.041</td>
<td>0.427</td>
<td>226.671</td>
<td>2</td>
</tr>
</tbody>
</table>
The solutions in the first category go from period $\tau = 0.413$ time units for $\nu = 0.054$ increasing monotonically until period $\tau = 0.484$ time units for $\nu = 0.049$. In the second category, the solutions go from period $\tau = 0.485$ for $\nu = 0.048$ decreasing monotonically until $\tau = 0.427$ for $\nu = 0.041$; for details concerning this, see Table 5.4. What we observe here is that when the solutions change from one maximum and one minimum for $\nu = 0.049$ to two maxima and two minima for $\nu = 0.048$, there is no big change in period, in fact, the difference is $10^{-3}$. An explanation for what is happening is that for $\nu = 0.049$ the solution is close to bifurcating and for $\nu = 0.048$ the solution is just beyond its bifurcation and changing slowly. In Figure 5.5 we compare the energy phase-planes of both cases; i.e., for $\nu = 0.049$ and $\nu = 0.048$. We have also checked the smoothness of this bifurcation by considering values of $I_E$ at several values of $\nu$ between 0.048 and 0.049. These calculations confirm the bifurcation structure described above.

![Figure 5.5](image)

**Figure 5.5** Comparison of energy phase-planes for $\nu = 0.049$ and $\nu = 0.048$. Here, $\eta = 1$, $\beta = 1$, $\Gamma_0 = 1$, $H_0$ in continuation format, $\Delta t = 10^{-3}$, $n = 128$, and time = 1000.

### 5.5 Tetra-Modal Traveling Attractors: $0.014 \leq \nu \leq 0.04$

It is found that for this interval, the solutions are attracted to tetra-modal ($\pi/2$-periodic) traveling waves. For the subinterval $0.028 \leq \nu \leq 0.04$, the wave speed
decreases monotonically as \( v \) decreases. It decreases from approximately \( c = 0.251 \) for \( v = 0.04 \) to approximately \( c = 0.0185 \) for \( v = 0.028 \) and the wave is traveling forward. Also, for this sub-interval, the interface energy, \( E_H \), has a convex shape. From the next value of \( v \), in parameter space, the wave speed decreases to negative values. So, for the sub-interval \( 0.014 \leq v \leq 0.027 \), the wave speed decreases monotonically as \( v \) decreases, ranging from approximately \(-0.0244\) for \( v = 0.027 \) to approximately \(-4.375\) for \( v = 0.014 \), and traveling backwards. The energy, in this sub-interval, is concave near the right end (\( v = 0.027 \)). Near the left end of this sub-interval, the energy increases with the feature that the rate of growth increases as we approach \( v = 0.014 \). For a physical interpretation of the relative positions of the three plotted profiles, see §5.1.

**Table 5.5** Wave speed (\( c \)) and interface energy (\( E_H \)) for \( 0.014 \leq v \leq 0.04 \). This is the tetra-modal traveling case. Here, \( \eta = 1 \), \( \Gamma_0 = 1 \), \( H_0 \) in continuation format, \( \Delta t = 10^{-3} \), \( n = 128 \), and \( \text{time} = 100 \).

<table>
<thead>
<tr>
<th>( v )</th>
<th>0.040</th>
<th>0.038</th>
<th>0.036</th>
<th>0.034</th>
<th>0.032</th>
<th>0.030</th>
<th>0.028</th>
</tr>
</thead>
<tbody>
<tr>
<td>( c )</td>
<td>0.251</td>
<td>0.222</td>
<td>0.195</td>
<td>0.167</td>
<td>0.130</td>
<td>0.081</td>
<td>0.018</td>
</tr>
<tr>
<td>( E_H )</td>
<td>488.518</td>
<td>495.226</td>
<td>495.967</td>
<td>490.727</td>
<td>479.529</td>
<td>462.471</td>
<td>439.840</td>
</tr>
<tr>
<td>( \nu )</td>
<td>0.027</td>
<td>0.024</td>
<td>0.022</td>
<td>0.020</td>
<td>0.018</td>
<td>0.016</td>
<td>0.014</td>
</tr>
<tr>
<td>( c )</td>
<td>-0.024</td>
<td>-0.240</td>
<td>-0.590</td>
<td>-1.375</td>
<td>-2.450</td>
<td>-3.400</td>
<td>-4.375</td>
</tr>
<tr>
<td>( E_H )</td>
<td>426.654</td>
<td>383.258</td>
<td>364.132</td>
<td>403.355</td>
<td>531.871</td>
<td>664.482</td>
<td>753.396</td>
</tr>
</tbody>
</table>

**5.6 Time Periodic Attractors: 0.011 \leq v \leq 0.013**

Once more, a time periodic attractor is entered. In this case, we use in our runs the values of \( \eta = 1 \), \( \Gamma_0 = 1 \), \( H_0 \) in continuation format, \( \Delta t = 10^{-3} \), and \( \text{time} = 1000 \), but 512 nodes in space. Using 128 or 256 nodes are not enough to perform successfully the Fast Fourier Transform (FFT) Method.
Figure 5.6 Profiles of interface (H), concentration of surfactant (Γ), and axial velocity (W₂) for η = 1, β = 1, Γ₀ = 1, H₀ in continuation format, Δt = 10⁻³, n = 128, and time = 100. For (a) ν = 0.034, c = 0.1670. For (b) ν = 0.021, c = -0.9035.

Figure 5.7 Energy phase-plane for ν = 0.012 in the third window of time periodic attractors 0.011 ≤ ν ≤ 0.013. Here, η = 1, Γ₀ = 1, H₀ in continuation format, Δt = 10⁻³, n = 128, and time = 1000.
All the cases in this interval have two maxima and two minima. In Figure 5.7, we plot the energy phase-plane for \( \nu = 0.012 \) in which we can clearly see the two maxima and two minima. Also, we observe that there is no significant change in period. It varies from \( \tau = 0.1504 \) at \( \nu = 0.013 \) to \( \tau = 0.1547 \) at \( \nu = 0.011 \), see Table 5.6 for details.

### Table 5.6
Period (\( \tau \)) and number of maxima and minima for \( \nu = 0.013, 0.012, \) and 0.011.

<table>
<thead>
<tr>
<th>( \nu )</th>
<th>( \tau )</th>
<th>Max./min.</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.013</td>
<td>0.1504</td>
<td>2</td>
</tr>
<tr>
<td>0.012</td>
<td>0.1540</td>
<td>2</td>
</tr>
<tr>
<td>0.011</td>
<td>0.1547</td>
<td>2</td>
</tr>
</tbody>
</table>

#### 5.7 Octa-Modal Traveling Attractors: \( 0.00375 \leq \nu \leq 0.01 \)
Similar to the tetra-modal case, the solutions are attracted to octa-modal (\( \pi/4 \)-periodic) travelling waves. Here, as in the bi-modal §5.3 and tetra-modal §5.5 cases we use \( \eta = 1, \Gamma_0 = 1, H_0 \) in continuation format, \( \Delta t = 10^{-3} \), and time = 100, but in all the cases we consider 512 nodes in space, except for \( \nu = 0.00375 \) in which we utilize 1024 space nodes. This is done in order to have sufficient Fourier modes in resolving sharp features and obtaining stable numerical solutions. Observing the data in Table 5.7, we can classify the solutions into two categories. As in the fully modal §5.1 and tetra-modal §5.5 cases, one category consists of forward-travelling solutions; i.e., the wave speed \( (c) \) positive, and the other category consists of waves which travel backwards; i.e., wave speed \( (c) \) negative. See Table 5.7 for details.

In the first category, \( 0.007 \leq \nu \leq 0.01 \), the wave speed decreases monotonically from the value \( c = 0.256 \) for \( \nu = 0.01 \) to the value \( c = 0.018 \) for \( \nu = 0.007 \). The wave is traveling forward and the energy, \( E_H \), is convex, i.e. initially the energy increases
Table 5.7 Wave speed \((c)\) and interface energy \((E_H)\) for \(0.00375 \leq \nu \leq 0.01\). Case of octa-modal traveling waves. Here, \(\eta = 1, \Gamma_0 = 1, H_0\) in continuation format, \(\Delta t = 10^{-3}\), \(time = 100\), and \(n = 512\) for all values of \(\nu\) except \(\nu = 0.00375\) in which we use \(n = 1024\).

<table>
<thead>
<tr>
<th>(\nu)</th>
<th>0.010</th>
<th>0.009</th>
<th>0.008</th>
<th>0.007</th>
</tr>
</thead>
<tbody>
<tr>
<td>(c)</td>
<td>0.256</td>
<td>0.155</td>
<td>0.140</td>
<td>0.018</td>
</tr>
<tr>
<td>(E_H)</td>
<td>1955.531</td>
<td>1982.739</td>
<td>1911.165</td>
<td>1739.050</td>
</tr>
</tbody>
</table>

and then decreases. In the second category, \(0.00375 \leq \nu \leq 0.006\), the wave speed decreases from \(c = -0.238\) at \(\nu = 0.006\) to \(c = -8.1\) at \(\nu = 0.00375\). In this sub-interval and starting from \(\nu = 0.006\), the energy decreases until we reach \(\nu = 0.005\), after which the energy starts to grow rapidly, see Table 5.7.

![Profiles of interface (H) and concentration of surfactant (\(\Gamma\))]({})

**Figure 5.8** Profiles of interface \((H)\), and concentration of surfactant \((\Gamma)\) for \(\eta = 1, \beta = 1, \Gamma_0 = 1, H_0\) in continuation format, \(\Delta t = 10^{-3}\), \(n = 512\), and \(time = 100\). For (a) \(\nu = 0.009, c = 0.1550\). For (b) \(\nu = 0.0045, c = -4.7\).

In Figure 5.8(a)(b), we plot the interface, \((H)\), and concentration of surfactant \((\Gamma)\) profiles for \(\nu = 0.009\) and \(\nu = 0.0045\), respectively. As in the other cases of
traveling wave behavior (§5.1, §5.3, and §5.5) the peaks in surfactant concentration are to be found ahead of the interfacial wave maximum. See §5.1 for a physical explanation of the relative positions of $H$, and $\Gamma$ profiles.

5.8 Time Periodic Attractors: $0.00275 \leq \nu \leq 0.0035$

In this window, $0.00275 \leq \nu \leq 0.0035$, the solutions are attracted to time periodic traveling waves. These solutions emerge as large time evolutions of our problem. For the computations, we use 1024 points in space, $\eta = 1$, $\Gamma_0 = 1$, $H_0$ in continuation format, $\Delta t = 10^{-3}$, and $time = 1000$. Here we found two types of behavior. Solutions with one value for the energy maxima and one for the energy minima, and solutions with two values for maxima and two for minima in the energy, $(E_H)$, profile. See Table 5.8 for more details.

Table 5.8 Period ($\tau$), integral of energy ($I_E$), and number of maxima and minima for $\nu = 0.0035, 0.00325, 0.003, \text{ and } 0.00275$.

<table>
<thead>
<tr>
<th>$\nu$</th>
<th>$\tau$</th>
<th>Integral of Energy ($I_E$)</th>
<th>Max./min.</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00350</td>
<td>0.0378</td>
<td>2684.071</td>
<td>1</td>
</tr>
<tr>
<td>0.00325</td>
<td>0.0452</td>
<td>3344.705</td>
<td>2</td>
</tr>
<tr>
<td>0.00300</td>
<td>0.0463</td>
<td>3863.520</td>
<td>2</td>
</tr>
<tr>
<td>0.00275</td>
<td>0.0501</td>
<td>4249.154</td>
<td>2</td>
</tr>
</tbody>
</table>

For $\nu = 0.0035$ the solution has one value for maxima and one for minima with period $\tau = 0.0378$. For the case of solutions with two values for maxima and two for minima, the period goes from $\tau = 0.0452$ for $\nu = 0.00325$ and increases monotonically until $\tau = 0.0501$ for $\nu = 0.00275$. In Figure 5.9, we have plotted the energy phase plane of both cases.
Figure 5.9  Energy phase-plane for (a) \( \nu = 0.0035 \) and (b) \( \nu = 0.00325 \) in the fourth window of time periodic attractors \( 0.00275 \leq \nu \leq 0.0035 \). Here, \( \eta = 1, \Gamma_0 = 1, H_0 \) in continuation format, \( \Delta t = 10^{-3}, n = 1024, \) and \( \text{time} = 1000. \)

5.9 Decahexa-Modal Traveling Attractors: \( 0.0009 \leq \nu \leq 0.0025 \)

As we expected, the solutions are attracted to decahexa-modal \( (\pi/8\)-periodic) traveling waves. In our simulations, we use 1024 nodes in space, the values of \( \eta = 1, \Gamma_0 = 1, H_0 \) in continuation format, \( \Delta t = 10^{-4}, \) and \( \text{time} = 15. \) As in the fully modal, tetra-modal, and octa-modal cases, §5.1, §5.5, §5.7, respectively, we have classified the solutions in two categories. The first one for those solutions that travel forward; i.e., wave speed \( (c) \) positive, and another category for those that travel backward; i.e., wave speed \( (c) \) negative. In Table 5.9, we include the wave speed, \( c, \) and energy, \( E_H \) of the solutions for eight different values of the parameter \( \nu. \)

The first category corresponds to the subinterval \( 0.00175 \leq \nu \leq 0.0025, \) where the wave speed decreases monotonically as \( \nu \) decreases. The wave speed goes from \( c = 0.280 \) for \( \nu = 0.0025 \) to \( c = 0.0195 \) for \( \nu = 0.00175. \) The second category is for solutions in the subinterval \( 0.0009 \leq \nu \leq 0.0015, \) where the wave speed decreases rapidly as \( \nu \) decreases, from \( c = -0.242 \) for \( \nu = 0.0015 \) to \( c = -16.9 \) for \( \nu = 0.00009. \)

In Figure 5.10(a)(b), we plot the interface, \( (H), \) and concentration of surfactant \( (\Gamma) \) profiles for \( \nu = 0.00225 \) and \( \nu = 0.00125, \) respectively. For a physical explanation
Table 5.9  Wave speed \( (c) \) and interface energy \( (E_H) \) for \( 0.0009 \leq \nu \leq 0.0025 \). Case of decahexa-modal traveling waves. Here, \( \eta = 1, \Gamma_0 \) and \( H_0 \) in continuation format, \( \Delta t = 10^{-3} \), \( \text{time} = 100 \), and \( n = 1024 \).

<table>
<thead>
<tr>
<th>( \nu )</th>
<th>0.00250</th>
<th>0.00225</th>
<th>0.00200</th>
<th>0.00175</th>
</tr>
</thead>
<tbody>
<tr>
<td>( c )</td>
<td>0.280</td>
<td>0.175</td>
<td>0.130</td>
<td>0.020</td>
</tr>
<tr>
<td>( E_H )</td>
<td>7749.028</td>
<td>7929.619</td>
<td>7637.532</td>
<td>6935.852</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>( \nu )</th>
<th>0.00150</th>
<th>0.00125</th>
<th>0.00100</th>
<th>0.0009</th>
</tr>
</thead>
<tbody>
<tr>
<td>( c )</td>
<td>-0.242</td>
<td>-2.503</td>
<td>-14.500</td>
<td>-16.900</td>
</tr>
<tr>
<td>( E_H )</td>
<td>5806.287</td>
<td>4521.286</td>
<td>9273.580</td>
<td>10592.740</td>
</tr>
</tbody>
</table>

Figure 5.10  Profiles of interface \( (H) \), and concentration of surfactant \( (\Gamma) \) for \( \eta = 1, \beta = 1, \Gamma_0 = 1, H_0 \) in continuation format, \( n = 1024 \), and \( \text{time} = 100 \). For (a) \( \nu = 0.00225, \Delta t = 10^{-3}, \) and \( c = 0.2 \). For (b) \( \nu = 0.00125, \Delta t = 10^{-4}, \) and \( c = -2.503 \).
of the relative positions of the interface, \((H)\), and concentration of surfactant \((\Gamma)\) profiles, see §5.1.

### 5.10 Time Periodic Attractors: \(0.000705 \leq \nu \leq 0.0008\)

In this interval, \(0.000705 \leq \nu \leq 0.0008\), a time periodic attractor is entered. In this case, we use in our runs the values of \(\eta = 1, \Gamma_0 = 1, H_0\) in continuation format, \(\Delta t = 10^{-3}\), and \(time = 1000\), but 2048 nodes in space to be able to obtain accurate solutions. Less than 2048 nodes were not enough to perform successfully the Fast Fourier Transform (FFT) Method.

![Energy phase-plane](image)

**Figure 5.11** Energy phase-plane for \(\nu = 0.000725\) in the fifth window of time periodic attractors \(0.000705 \leq \nu \leq 0.0008\). Here, \(\eta = 1, \Gamma_0 = 1, H_0\) in continuation format, \(\Delta t = 10^{-3}\), \(n = 2048\), and \(time = 100\).

All the cases, in this interval, have two maxima and two minima. In Figure 5.11, we plot the energy phase-plane for \(\nu = 0.000725\) in which we can clearly see the two maxima and two minima. Also, we observe that there is little variation in the period. It starts from \(\tau = 0.0131\) at \(\nu = 0.0008\) to \(\tau = 0.0155\) for \(\nu = 0.000705\), see Table 5.10.
Table 5.10  Period (τ) and number of maxima and minima for $0.000705 \leq \nu \leq 0.0008$.

<table>
<thead>
<tr>
<th>$\nu$</th>
<th>$\tau$</th>
<th>Max./min.</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0008</td>
<td>0.0131</td>
<td>2</td>
</tr>
<tr>
<td>0.00075</td>
<td>0.0137</td>
<td>2</td>
</tr>
<tr>
<td>0.000725</td>
<td>0.0144</td>
<td>2</td>
</tr>
<tr>
<td>0.0007125</td>
<td>0.0150</td>
<td>2</td>
</tr>
<tr>
<td>0.00070625</td>
<td>0.0154</td>
<td>2</td>
</tr>
<tr>
<td>0.000705</td>
<td>0.0155</td>
<td>2</td>
</tr>
</tbody>
</table>
CHAPTER 6

CONCLUSIONS

This work has addressed the problem of the effect of surfactants on the nonlinear stability of core-annular flows. We have shown that an asymptotic theory can be used when the core fluid (a viscous fluid such as crude oil in applications) occupies most of the tube and is lubricated by a thin less viscous layer (usually water in applications involving oil transport). Starting from the Navier-Stokes equations in circular tubes, the axisymmetric two-fluid problem has been formulated in full generality and for arbitrary axisymmetric interfacial disturbances of order $O(\varepsilon^2)$, where $\varepsilon$ is the thickness of the film. Insoluble surfactants are present on the sharp interface separating the two phases, and the flow is driven by a constant axial pressure gradient which produces an undisturbed state of a perfectly cylindrical core being lubricated by a uniform thickness annulus. In the base state, the surfactant concentration is uniform implying a uniform surface tension and the resulting flow is a piecewise continuous Poiseuille flow.

The present work introduces a small parameter, $\varepsilon$, which is equal to the ratio of the undisturbed annulus thickness to the undisturbed core radius. Matched asymptotic expansions are used to derive a system of coupled nonlinear partial differential equations which, in scaled terms, represent the evolution of the interfacial amplitude separating core and film fluids, and the evolution of interfacial surfactant concentration. In the absence of surfactants, the equation reduces to the Kuramoto-Sivashinsky equation. The effects of viscosity stratification have also been retained, leading to a dispersive modification of the Kuramoto-Sivashinsky equation through a pseudo-differential term.

The second aspect of this dissertation is concerned with the numerical solution of the derived evolution equations, and in particular classification of the nonlinear
dynamics as well as evaluation of the effect of surfactants on the system. The calculations consider the dynamics in the absence of dispersive effects, in which case the surfactant-free system is the Kuramoto-Sivashinsky equation which is known to possess a rich class of complicated and chaotic dynamics. Several numerical schemes have been tried and implemented and we choose an implicit second order accurate scheme based on the spectral representation of derivatives and pseudo-spectral representation of nonlinear terms. The code was tested against published results of the Kuramoto-Sivashinsky equation and the agreement is excellent.

Over 300 numerical experiments have been performed in order to piece together the following picture of the dynamics as the bifurcation parameter \( \nu = \frac{\pi^2}{L^2} \) where \( L \) is the size of the system) is varied in the range \((0, \infty)\); note that as \( \nu \to 0 \) the size of the system becomes infinite and the band of linearly unstable modes increases without bound. The case \( \nu = 0 \) gives a negative diffusion nonlinear parabolic equation and so the problem is ill-posed.

Two types of motion have been established: (i) Nonlinear traveling waves for most values of \( \nu \), (ii) Time-periodic traveling waves in small windows separating traveling wave attractors. For relatively large values of \( \nu \) (larger than about 0.2) fully modal traveling waves are seen. The amplitudes and speed of the waves tends to zero as \( \nu \) becomes large. This is in contrast to the surfactant-free system where the solutions for \( \nu > 1 \) tend to uniform trivial states. By fully-modal we mean that all Fourier coefficients of the periodic waves are non-zero.

The traveling waves have an interesting structure. First, in physical space all waves travel in the direction of the base flow. The surfactant concentration has a peak just ahead of the interfacial wave maximum and this sets up Marangoni forces away from this central region. The explanation for this lies in the perturbation axial velocity distribution obtained from the leading order asymptotic solution, which is found to sweep surfactant into the peak region until a nonlinear balance is achieved. This way,
the peak in surfactant concentration is slaved relative to the peak in the interfacial amplitude and in all of our simulations it is always in front of the latter. This delicate but at the same time general feature, could guide experimental observations.

As $\nu$ is decreased further, a time-periodic attractor is found in the range $0.099 \leq \nu \leq 0.19$. Three disjoint attractors have been established in this window which lead to the conclusion that one attractor loses stability and the solution jumps to a different branch with a different period as well as temporal structure. This has been established by monitoring a positive definite quantity that remains smooth on bifurcations such as subharmonic cascades, for instance. This particular window supports three separate sub-windows with different solution branches.

A decrease of $\nu$ into the range $0.055 \leq \nu \leq 0.099$ produces traveling waves which are $\pi$-periodic or bi-modal (by this we mean that only the even Fourier modes of the periodic waves are non-zero). As the value of $\nu$ is decreased, the waves increase in amplitude and the wave speeds become increasingly more negative as we exit the attractor.

On exit, we enter another time-periodic attractor over the range $0.041 \leq \nu \leq 0.054$. The time-period of the solutions ranges between 0.4 and 0.5, approximately, and supports two sub-windows with different temporal characteristics. The change is found to be smooth and so the same attractor is at play.

As $\nu$ is decreased further a $\frac{\pi}{2}$-periodic or tetramodal attractor (that the only non-zero Fourier coefficients of the periodic waves are multiples of 4) emerges. The energy $E_H$ defined elsewhere (this is a measure of the wave size), is large and the interfacial amplitudes become as large as 10-15 units.

The next window, $0.011 \leq \nu \leq 0.013$ is another time-periodic attractor supporting 2 energy maxima and minima and producing solutions on a single branch. The period is almost constant and approximately equal to 0.15. This feature, of
decreasing time period in successively lower time-periodic attractors, is found to be true in all our numerical calculations.

The next computed attractor is in the range $0.00375 \leq \nu \leq 0.01$ and supports octamodal traveling waves (i.e. waves of period $\frac{\pi}{4}$ whose only non-zero Fourier modes are multiples of 8). The wave speed decreases monotonically and is negative and equal to $-6.7$ at exit. In addition the amplitudes are large and of the order of 25-30 units.

Another time periodic attractor is found next, in the range $0.00275 \leq \nu \leq 0.0035$. The period varies between 0.0378 to 0.0501 and the wave amplitudes are large with the energy becoming as large as $4 \times 10^8$. These results were obtained with 1024 modes.

On further decrease of $\nu$, the most strongly attracting state is now a traveling wave attractor over the range $0.0009 \leq \nu \leq 0.0025$. The spatial wave period is $\frac{\pi}{8}$, and the attractor is decahexa-modal, i.e. the only non-zero Fourier coefficients of the waves are multiples of 16. The number of modes used is 1024 and the speed decreases monotonically to values as large as $-16.9$ on exit. The wave amplitudes are of the order of 50 units.

The final computed attractor (these computations are quite delicate and require at least 2048 modes) is found to be time-periodic and is in the range $0.000705 \leq \nu \leq 0.0008$. The time-period is decreased relative to the last periodic attractor and ranges between 0.0131 and 0.0155 time units. A single branch of solutions was found.

Even though we did not carry out calculations for smaller values of $\nu$, we have established a fairly robust pattern through our extensive simulations. The alternating behavior of time-periodic and multimodal traveling waves remains to be tested more thoroughly, but it is tempting to conclude that there is a succession of successively decreasing in size windows which support time-periodic solutions with successively decreasing periods. In addition, in between any two time-periodic windows, there
exists a traveling wave attractor which lives on successively decreasing window lengths and is periodic of period $\frac{2\pi}{2^n}$, where $n = 0, 1, 2, \ldots$. We have computed as far as $n = 4$. We have also established a self-similar relationship between solutions in these successively higher modal solutions and have confirmed such finding numerically.
In this appendix, we explain how we compute the curvature of the disturbed interface. In the Figures A.1 and A.2, for a better visualization, we have drawn the normal vector \( \vec{n} \) pointing out, but in our problem the normal vector \( \vec{n} \) is pointing in.

The curvature, \( \kappa \), is given by \( \kappa = \kappa_1 + \kappa_2 \), where \( \kappa_1 = \frac{1}{R_1} \) and \( \kappa_2 = -\frac{1}{R_2} \) with \( R_1 \) the radius of the circumference circumscript to the interface in a plane parallel to the interfacial normal vector \( \vec{n} \) with an angle \( \theta \) between the plane and the cylinder axis (see Figure A.1), and \( R_2 \) the radius of the circumference circumscript to the interface in a plane parallel to the cylinder axis (see Figure A.2).

Let us compute first the curvature \( \kappa_1 = \frac{1}{R_1} \). We define \( \vec{x} = S\vec{e}_r + z\vec{e}_z \) as in §2.2. The curvature of the circumference circumscript to the interface in a plane parallel to the interfacial normal vector is defined as the curvature of a plane curve and is given by

\[
\kappa_1 = \frac{\left| \vec{V} \times \vec{a} \right|}{\left| \vec{V} \right|^3},
\]

(A.1)

**Figure A.1** Radius \( (R_1) \) of the circumference circumscript to the interface in a plane parallel to the unit normal vector, \( \vec{n} \), and forming and angle \( \theta \) with the cylinder axis.
where $\mathbf{V} = \frac{\partial \mathbf{x}}{\partial z}$ and $\mathbf{a} = \frac{\partial \mathbf{V}}{\partial z}$, see Thomas and Finney ([75], 1992). Thus, we have

$$\mathbf{V} = (S', 0, 1) \quad \text{and} \quad \mathbf{a} = (S'', 0, 0);$$  \hspace{1cm} (A.2)

hence, $\mathbf{V} \times \mathbf{a} = S''$. Thus,

$$|\mathbf{V} \times \mathbf{a}| = S'' \quad \text{and} \quad |\mathbf{V}| = \sqrt{1 + (S')^2}$$  \hspace{1cm} (A.3)

Therefore,

$$\kappa_1 = \frac{S''}{(1 + (S')^2)^{3/2}}.$$  \hspace{1cm} (A.4)

Now, we compute $\kappa_2 = \frac{1}{R_2}$. The radius $R_2$ of the circumference circumscribed to the interface in a plane parallel to the cylinder axis is given by

Figure A.2  Radius ($R_2$) of the circumference circumscribed to the interface and parallel to the cylinder axis. $\mathbf{n}$ is the unit normal vector and $\mathbf{t}$ is the tangential unit vector with slope $S'$, of the interface; and $\theta$ is the angle formed between $\mathbf{n}$ and the cylinder axis.

$$\kappa_2 = \frac{1}{R_2},$$  \hspace{1cm} (A.5)
where $R_2$ is the radius of the circumference circumscribed to the interface in a plane parallel to the cylinder axis; so

$$R_2 = \frac{S}{\sin \theta}, \quad (A.6)$$

where $\theta$ is the angle formed between the segment, on which the normal unit vector rest, and the axis of the cylinder. Looking at the triangle in Figure A.2, we see that

$$\sin \theta = \frac{1}{\sqrt{1 + (S')^2}}. \quad (A.7)$$

Therefore,

$$\kappa_2 = -\frac{1}{S \sqrt{1 + (S')^2}}. \quad (A.8)$$

Hence, from (A.4) and (A.8), we obtain

$$\kappa = \frac{S''}{(1 + (S')^2)^{3/2}} - \frac{1}{S \sqrt{1 + (S')^2}}$$

$$= \frac{1}{S \sqrt{1 + (S')^2}} \left[ \frac{SS''}{1 + (S')^2} - 1 \right]. \quad (A.9)$$
APPENDIX B

ORDER OF THE SCHEME (4.7) AND (4.8)

In this appendix, we give a proof of the $O((\Delta t)^2)$ accuracy of the scheme (4.7) and (4.8).

Let us show that the second system (B.2) is of $O(\Delta t^2)$.

Rewriting (B.2) in single equations, we have, for the $k$-th mode:

\[
\frac{\hat{H}^{t+\frac{1}{2}} - \hat{H}^t}{(\Delta t/2)} + M(k) \begin{bmatrix} \hat{H}^{t+\frac{1}{2}} \\ \hat{\Gamma}^{t+\frac{1}{2}} \end{bmatrix}^{(k)} = \begin{bmatrix} f_1^{t} \\ f_2^{t} \end{bmatrix}^{(k)}
\] (B.1)

\[
\begin{bmatrix} \frac{\hat{H}^{t+1} - \hat{H}^t}{\Delta t} \\ \frac{\hat{\Gamma}^{t+1} - \hat{\Gamma}^t}{\Delta t} \end{bmatrix}^{(k)} + \frac{1}{2} M(k) \begin{bmatrix} \hat{H}^{t+1} + \hat{H}^t \\ \hat{\Gamma}^{t+1} + \hat{\Gamma}^t \end{bmatrix}^{(k)} = \begin{bmatrix} f_1^{t+\frac{1}{2}} \\ f_2^{t+\frac{1}{2}} \end{bmatrix}^{(k)}
\] (B.2)

where $M(k) = \begin{bmatrix} \nu k^4 - k^2 & -k^2 \\ 0 & \eta k^2 \end{bmatrix}$.

The first system (B.1) is $O(\Delta t)$ accurate. Let us show that the second system (B.2) is of $O((\Delta t)^2)$. Rewriting (B.2) in single equations, we have, for the $k$-th mode:

\[
\frac{\hat{H}^{t+1} - \hat{H}^t}{\Delta t} + \frac{1}{2}(\nu k^4 - k^2)(\hat{H}^{t+1} + \hat{H}^t) - \frac{1}{2} k^2(\hat{\Gamma}^{t+1} + \hat{\Gamma}^t) = f_1^{t+\frac{1}{2}}
\] (B.3)

\[
\frac{\hat{\Gamma}^{t+1} - \hat{\Gamma}^t}{\Delta t} + \frac{1}{2} \eta k^2(\hat{\Gamma}^{t+1} + \hat{\Gamma}^t) = f_2^{t+\frac{1}{2}}
\] (B.4)

Expanding $\hat{H}^{t+1}$, $\hat{\Gamma}^{t+1}$, $f_1^{t+\frac{1}{2}}$, and $f_2^{t+\frac{1}{2}}$ about $t$, and simplifying terms, we get:

\[
\hat{H}^t + \frac{1}{2}(\Delta t)\hat{H}_{tt}^t + \frac{1}{6}(\Delta t)^2\hat{H}_{ttt}^t = (\Box) - (\Box) + (\Box),
\]

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where
\[
\Box = \left( f_1^t + \frac{1}{2}(\Delta t)f_{1t}^t + \frac{1}{8}(\Delta t)^2 f_{1tt}^t + \ldots \right) \\
\Diamond = (\nu k^4 - k^2)\hat{H}_t^t + \frac{1}{2}(\nu k^4 - k^2)\Delta t \left[ \hat{H}_t^t + \frac{1}{2}(\Delta t)\hat{H}_{tt}^t + \ldots \right] \\
\bigcirc = k^2\hat{f}_t^t + \frac{1}{2}k^2(\Delta t) \left[ \hat{f}_t^t + \frac{1}{2}(\Delta t)\hat{f}_{tt}^t + \ldots \right],
\]

and
\[
\hat{f}_t^t + \frac{1}{2}(\Delta t)\hat{f}_{tt}^t + \frac{1}{6}(\Delta t)^2\hat{f}_{ttt}^t = (\Box) + (\Delta),
\]

where
\[
\Box = \left( f_2^t + \frac{1}{2}(\Delta t)f_{2t}^t + \frac{1}{8}(\Delta t)^2 f_{2tt}^t + \ldots \right) \\
\triangle = \eta k^2\hat{f}_t^t + \frac{1}{2}\eta k^2(\Delta t) \left[ \hat{f}_t^t + \frac{1}{2}(\Delta t)\hat{f}_{tt}^t + \ldots \right].
\]

The terms of \(O(1)\) are automatically satisfied, see (4.4) and (4.5). Taking the \(O(\Delta t)\) terms, we have that
\[
\hat{H}_{tt}^t + (\nu k^4 - k^2)\hat{H}_t^t - k^2\hat{f}_t^t = f_{1t}^t \\
\hat{f}_t^t + \eta k^2\hat{f}_t^t = f_{2t}^t.
\]

But this is just the time derivative of (4.4) and (4.5). Now, taking the \(O((\Delta t)^2)\) terms, we find the following
\[
\hat{H}_{ttt}^t + \frac{3}{2}(\nu k^4 - k^2)\hat{H}_{tt}^t - \frac{3}{2}k^2\hat{f}_{tt}^t = \frac{3}{4} f_{1tt}^t \\
\hat{f}_{tt}^t + \frac{3}{2}\eta k^2\hat{f}_{tt}^t = \frac{3}{4} f_{2tt}^t,
\]

and these are not satisfied by (4.4) and (4.5). Therefore, the system (B.2) is of \(O((\Delta t)^2)\).
APPENDIX C

DIAGNOSTIC OF ATTRACTORS

In this appendix, we give a definition of an attractor, and diagnostics of the type of attractors. An **attractor** is an invariant set $A$ for which in some neighborhood $U$ of $A$ such that: $H(v; 0)$ in $U$ implies $H(v; t)$ in $U$ for all $t > 0$, and $H(v; t) \rightarrow A$ as $t \rightarrow \infty$ (Perko [57], 1998). The energy of the solution in $L^2$ is defined by the $L^2$-norm as

$$E(t) = \int_0^{2\pi} u^2(z, t) dz,$$

which can be computed along with the solution using the trapezoidal rule

$$E(t) = \Delta z \sum_{j=1}^{n} u_j^2(t),$$

where $u_j(t) \equiv u(z_j, t)$, and $z_j = j\Delta z$ for $j = 0, \ldots, n - 1$. We note that trapezoidal quadrature is spectrally accurate for periodic functions.

We are interested in the behavior of the energy since its time evolution is illustrative of the nature of various attractors. If the energy $E(t)$ evolves to a non-zero constant state as time increases then the solution evolves to a stationary or traveling steady state. The interface profile and the concentration of surfactant profile are helpful in classifying an attractor as **stationary** or a **steady-state traveling** wave by comparing the profiles for two different times. In the case of a space-periodic traveling wave attractor, we can compute the wave speed using the profiles for those two different times, once transients have died out and the solution reaches its long time behavior.

Time-periodic, quasi-periodic, and chaotic attractors evolve to an oscillatory state of energy in time. A useful way to confirm whether a solution evolves to a **time-periodic** attractor is to numerically construct the energy phase-plane $(E, dE/dt)$. 

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If the phase-plane is a closed curve, then the solution evolves to a time-periodic attractor. The index of the closed curve which is the number of points where the phase curves intersect the $E$-axis, i.e. $dE/dt = 0$, determines the number of maxima and minima of $E(t)$ in one period. In the cases when it is not easy to detect how many maxima and minima the energy has in one period, we can use the return map for maxima and for minima. If $\{a_n\}$, $n$ a natural number, is a sequence of real numbers, the **Return Map** of $\{a_n\}_{n=0}^{\infty}$ is the set of points $(a_n, a_{n+1})$, $n$ a natural number. The number of points in the return map, after transients die out, is the number of maxima (or minima). The index can also be used to follow possible period-doubling bifurcations. However, we like to have a more accurate period to better determine period doubling bifurcations. In order to do so, we use second order polynomial interpolation over a very large number of points $(t_j, E(t_j))$.

There are cases where it seems that there is period doubling, but really it is not, see §5.2. A smooth bifurcation would result in the case of a period doubling, for example. Instead, what is happening in §5.2 is that one periodic state becomes unstable to a different coexisting periodic state. One way to quantify this is to consider the integral $I_E(\nu, \tau) = \frac{1}{\tau} \int_{t_0}^{t_0 + \tau} E(t; \nu) dt$, where $\tau$ is the period of the oscillation and $t_0$ is an arbitrary time chosen so that the transients have died out. If the values of $I_E$ of the solutions from one interval are very different from the ones in another interval, then the solution branches are not smoothly connected. An example of that is found in Table 5.2 where inspection of the values of $I_E$ shows that the one, three, and five maxima/minima solution branches are not smoothly connected, even though looking at the values of $\tau$ it appears that there may be a period doubling.
BIBLIOGRAPHY


