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Interfacial Dynamics of Lamellar Phases

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Abstract

A lamellar phase is a lyotropic smectic A liquid crystal, composed of parallel bilayers of surfactant separated by water. Lamellar phases and states of coexistence between lamellar phases and isotropic phases form significant parts of the phase diagrams of surfactant solutions.

In this work we examined the dynamics of interfaces between a smectic A, and in particular a lamellar phase, and a simple fluid or gas, at low Reynolds numbers.

Although surface modes of thermotropic smectic A are governed by inertia, we have shown that for lamellar phases, modes dominated by surface tension and elasticity play an important role. These modes become relevant due to an additional dissipative mechanism unique to lyotropic lamellar phases, the slip mechanism, allowing relative motion of the surfactants and water layers.

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1 Introduction

The field of liquid crystals began to be seriously researched only fifty years ago. These visco-elastic materials have unique hydrodynamic properties. They are able to flow, as liquids, and they also have inner symmetries (which is a crystal feature) that enable them to generate elastic responses.

Smectic A is a liquid crystal composed of parallel liquid layers, one on top of the other. Lamellar phase is a smectic A liquid crystal in which the layers are composed of surfactant molecules (bilayer membranes), and the gap between them is occupied by water.

Smectic phases and specifically lamellar phases are widespread in the cosmetics and cleaning industry, in products such as soaps and shampoos due to the surfactant ability to connect water and oil (indeed; the term smectic comes from the Greek word for soap.)

A similar structure, the multi-lamellar vesicle, is composed of up to thousands of spherical bilayers, forming a spherical lamellar phase. Such structures are used in the pharmaceutical industry. Bilayer membrane structures form the basic envelope in biological systems as cells and vesicles (the latter play an important role in intra-cellular mobilization.) Complex intra-cellular structures like the mitochondria also show a multi-bilayer composition. Another liquid crystal - the nematic liquid crystal, has allowed major progresses in the field of Liquid Crystal Display (LCD).

In this work we studied the hydrodynamics of surface modes forming between smectic A liquid crystal or a lamellar phase and gas or a simple fluid. Surface modes are perturbations on the interface, decaying at increasing distances from it. The bulk equations for the phases' hydrodynamics yield the dependence of the spatial decay on the temporal decay, and the relation between the hydrodynamic functions. The boundary conditions determine completely the eigen modes for the dynamics.

A similar approach for defining the surface modes was employed in a former study by Fedorov et al [1], dealing with smectic A ruled by inertia. Fedorov et al's research showed that one inertia-dominated surface mode of the smectic A is a Rayleigh wave (surface acoustic wave) of the second sound type: $\omega = \pm(\sqrt{B/\rho})q - i\omega''$ where B is the compression modulus

and ρ is the mass density. $i\omega$ and q are the temporal decay and the wave number respectively. ω'' gives the temporal decay. This mode exists in the area of $w \ll B/\eta_3$ and $w^2 \simeq (B/\rho)q^2$, where η_3 is one of the viscosity coefficient. Fedorov et al [1] describe also a second surface mode: $\omega = -4i\eta_3q^2/(3\rho)$ which appears in the limit $\omega \gg B/\eta_3$ or $\omega^2 \gg (B/\rho)q^2$. This is a diffusive mode ($\omega \propto q^2$).

The "Orsay group on liquid crystals" [2] suggested that for thick samples of smectic material, the surface modes would be the same as for low Reynolds number simple fluid with renormalized surface tension, as known from static calculations $\tilde{\gamma} = \gamma + \sqrt{BK}$, where γ is the surface tension coefficient and K is the bending modulus. Meaning $\omega = -i(\gamma + \sqrt{BK})/2\tilde{\eta}$, where $\tilde{\eta}$ represent a characteristic viscosity. Their mode, though, does not fulfill the boundary conditions at the interface.

There is no previous study on the surface modes that are unique to lamellar phases. In this research we propose an asymptotic description for these surface modes. The current state of knowledge lacks also a full description for smectic A surface tension and elasticity dominated surface modes. We propose a solution for both questions in this research. Furthermore, we compare the lamellar phase modes to the surface modes of simple fluid.

The systems studied are at equilibrium. Therefore, a small fluctuation at the interface decays back to zero. The temporal decay of smectic A surface modes is influenced (neglecting inertia) by surface tension and the two elastic features of bending and compressibility, that characterize the smectic phase.

For a single-component smectic A we have found that when inertia is negligible the dominated surface mode resembles that of simple fluid. In this mode the temporal decay depends linearly on the wave number, and is influenced mostly by the surface tension. The spatial decays also have a linear dependence on the wave number (we refer to temporal decay as a parameter with the dimension of frequency, and to spatial decay as a parameter with the dimension of 1/length.) The smectic viscosity is anisotropic. Therefore, several viscosity coefficients are needed to characterize this phase. Each mode exhibits a different typical viscosity. However, we find that these zero-Reynolds-number modes are valid over a very restricted range of wavelengths, thus confirming the conclusion of Ref [1] as to the

dominance of inertial surface modes in these single-component phases.

For lamellar phases, there is an extra degree of freedom (compared with smectic A), since the lamellar phase is a two component fluid [3]. This property is called slip, it represents the possible existence of relative velocity between the surfactants and the water in the direction parallel to the layers. The slip property changes the possible surface modes and enlarges the relevant range of wave lengths ruled by surface tension and elastic qualities. The modes influenced by the slip property in the lamellar phase have decay rates that depend quadratically on the wave number, in contradistinction with the surface modes appearing in smectic A, where the decay rate shows linear dependence on the wave number.

A description of the lamellar phase, its hydrodynamics and interfaces' boundary conditions are presented in the following background section. The specific boundary conditions for interfaces between smectic A or a lamellar phase and a simple fluid or gas are discussed in section 3. For smectic A dimensional analysis can give roughly the possible decay rates as we show in section 4. In sections 5, 6 and 7 we calculate these modes specifically for general smectic A. In sections 8 and 9 we address the influence of the slip property on the lamellar phase surface modes.

2 Background

2.1 What is a Lyotropic Lamellar Phase

Macro - How Can a Liquid be a Crystal ?

A lyotropic ¹ lamellar phase is a liquid crystal. To be specific, it is a two-component smectic A liquid crystal. The name “liquid crystal” seems self-contradicting: how can a liquid be a crystal ?

A liquid is a disordered material, a feature which is essential for it to be able to flow (fluid). A crystal is an ordered material. Order means that the atomic structure has symmetries. This fixation of the atoms keeps the material as a solid, and it cannot flow. Liquid crystals are liquids in the sense that they are able to flow. Nevertheless they have an inner symmetry, which is a feature of crystals. This crystal behavior will yield elastic forces, and anisotropy.

Micro - The Microscopic Structure of a Lyotropic Lamellar Phase

The lyotropic lamellar phase is a two component fluid. It is composed of water molecules and surfactant molecules.

Surfactant is an amphiphilic molecule containing a hydrophilic “head” and a hydrophobic “tail”. Low energy states of a system containing these molecules are achieved when the surfactant heads are in polar solution, and the “tails” are in oily surrounding.

In an aqueous solution whose boundaries are with air or oil, surfactant molecules will gather on the interface. Sufficient amount of surfactant molecules in the solution causes the surfactants to associate together in structures depending on their concentration, the relative sizes of “head” and “tail”, temperature, electric forces etc. The surfactants can associate into micelles or a variety of bilayer structures.

A micelle is a cluster made of surfactant molecules where the hydrophilic heads are oriented toward the surrounding water, and the hydrophobic tails are oriented toward the

¹lyotropic phase is composed of two components whereas thermotropic phase consists of only one component.

center. Surfactant molecules can also form bilayers where heads are oriented toward the outward of the layer and tails are oriented inside. The bilayers are also referred to as “membranes”. The surfactant molecules maintain a two dimensional fluid on the bilayer surface.

The bilayers can form a variety of structures: vesicles (spheres), sponge phases (multiply-connected, sponge-like film), lamellar phases (roughly flat, bilayer sheets forming a stack – a type of a smectic A liquid crystal described further), and also onion phase, hexagonal smectic A, smectic B, smectic C, etc [4] [5].

Smectic liquid crystals are anisotropic multi-layer structures, where the layers are parallel on average and each layer acts as a two dimensional fluid. In smectic A the translation symmetry is broken in the direction perpendicular to the layers. Lyotropic lamellar phase is a smectic A liquid crystal made of bilayers of surfactant molecules, where the layers are separated by water molecules (Fig 1). The thickness of the water layers vary between 5 Å in un-charged lipids to 250 Å for charged lipids [3].

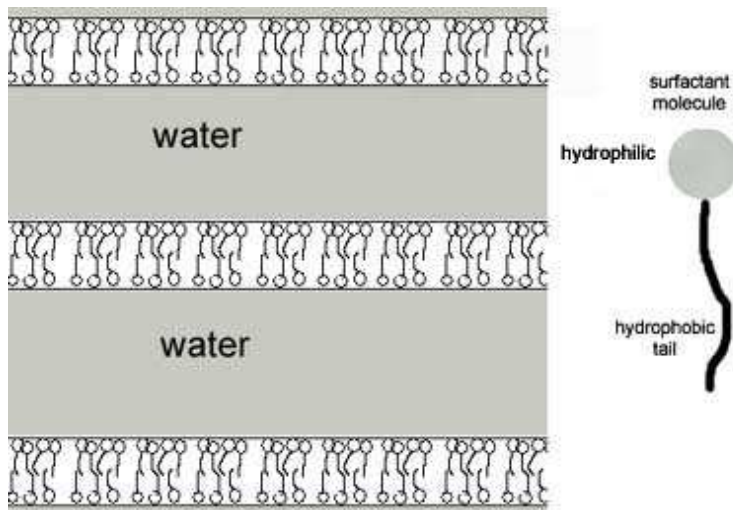


Figure 1: Lamellar Phase

The spacing between lamellar phase bilayers d is determined by concentration, Van-Deer-Waals attraction, Helfrich repulsion, electric repulsion and hydration repulsion forces. Helfrich repulsion is a steric repulsion arising from the reduction in the layers’ undulation

entropy as their spacing decreases [6].

The hydrodynamic properties of a lamellar phase have, besides the features of simple fluids, other unique features:

- The attraction and repulsion interactions between layers induce an elastic compression modulus.
- The elasticity of each bilayer membrane contributing to an elastic bending modulus for the entire phase.
- Anisotropy of the viscosity tensor.
- Permeation of matter through the layers.
- Normal diffusion - movement of the surfactant between layers.
- Slip mode due to relative lateral motion of the water and the surfactant.

The first four properties are common to any smectic A. The fifth and the sixth properties coincide for thermotropic smectics. The last property, the slip mode, is relevant specifically for lamellar phases, since it characterizes a two component smectic.

2.2 General Hydrodynamic Equations Relevant to the Study

Continuity

The continuity equation is constructed from the principle of mass conservation. The mass of the material bounded inside an arbitrary volume changes in time according to the net flux through that volume.

$$\iiint \frac{\partial \rho}{\partial t} dV = - \iint \mathbf{j} \cdot d\mathbf{S} \quad (1)$$

ρ is the mass density, \mathbf{j} is the momentum flux, V and \mathbf{S} represents volume and surface respectively, where \mathbf{S} is directed normal to the surface.

The differential representation of this principle is:

$$\frac{\partial \rho}{\partial t} = -\nabla \cdot \mathbf{j} \quad (2)$$

Navier-Stokes

Navier-Stokes Equations result from Newton's second law. Its integral form, for a volume element, is:

$$\iiint \rho \frac{d\mathbf{v}}{dt} dV = \iint \mathbf{t} dS + \iiint \rho \mathbf{b} dV \quad (3)$$

\mathbf{t} is a contact force acting on the element surface. \mathbf{b} is a force operating without contact (For example: \mathbf{t} can be pressure or viscosity and \mathbf{b} can be a body force such as gravitation.)

From Cauchy theorem \mathbf{t} is homogeneous and linear in the surface's normal \mathbf{n} :

$$\mathbf{t}(\mathbf{x}, t) = \sigma(\mathbf{x}, t)\mathbf{n} \quad (4)$$

The matrix σ is called the stress tensor, it is independent of the surface orientation. The direction of the force is set by \mathbf{n} alone. σ_{ij} is the force in the direction \mathbf{i} acting on a surface whose normal is in the \mathbf{j} direction.

According to the divergence theorem, the two dimensions integral can be written as a volume integral. Therefore, for continues media differential representation of Navier-Stokes' equation is:

$$\rho \left(\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{v} \right) = \nabla \cdot \sigma + \rho \mathbf{b} \quad (5)$$

Where a matrix divergence is defined as: $(\nabla \cdot \sigma)_i = \sum_j \frac{\partial \sigma_{ij}}{\partial x_j}$. The left hand side results from dividing the complete time derivatives into partial derivatives with respect to time and position.

2.3 Simple Fluid Hydrodynamics

For constant temperature processes, the hydrodynamics are determined by mass conservation and Navier-Stokes equation. The Navier-Stokes equation has the general form presented in Eq. 5, where the stress tensor σ is composed of the isotropic pressure, and the viscous stress tensor:

$$\sigma'_{ij} = \eta \left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} - \frac{2}{3} \delta_{ij} \frac{\partial v_k}{\partial x_k} \right) + \zeta \delta_{ij} \frac{\partial v_k}{\partial x_k}$$

For incompressible fluid $\frac{\partial v_k}{\partial x_k} = 0$.

In this work small fluctuations on equilibrium are being considered. Therefore, the non linear term $(\mathbf{v} \cdot \nabla)\mathbf{v}$ is in second order in the perturbation and is hence omitted from Navier-Stokes equation.

If the force for mass unit \mathbf{b} is zero and the simple fluid is incompressible (as hereafter assumed), the hydrodynamic equations are simplified.

Mass Conservation:

$$\nabla \cdot \mathbf{v} = 0 \tag{6}$$

Navier-Stokes:

$$\rho \frac{\partial \mathbf{v}}{\partial t} = -\nabla p + \eta \nabla^2 \mathbf{v} \tag{7}$$

Reynolds Number

The expression for Reynolds number is developed here to be relevant for the surface mode problem investigated in this work. For a wave, a typical length scale is the wave length ($\lambda = 2\pi q^{-1}$). Let the typical time scale of the systems dynamics be $\tau = 2\pi\omega^{-1}$. Then, the flow is specified by the system's typical parameters ω , $\frac{\eta}{\rho}$ and q . Only one dimensionless quantity can be formed from the above three, this combination is called the Reynolds Number and is denoted by Re:

$$\text{Re} = \frac{i\rho\omega}{q^2\eta}$$

Presenting Navier-Stokes equation by dimensionless parameters, shows that the ratio between the inertial parts and the viscous part is the Reynolds number. Hence the inertial terms can be neglected for low Reynolds number. This is the case when we examine slowly decaying surface modes of lamellar phase. We shall use this in our derivation of the surface modes.

The low-Re hydrodynamics of simple fluids are well known. In writing the above part of the thesis I was helped by the book "Fluid Mechanics" by Landau and Lifshitz [7].

2.4 Lamellar Phase Hydrodynamics

The theory of smectic A hydrodynamics was constructed by Martin et al [8], and is described also in a clear form in the book "Theory of Elasticity" by Landau and Lifshitz [9], where, in

addition the elastic tensor is presented. The viscosity tensor used in the present work is from Brochard and de-Gennes [3], in which the authors specifically addressed the issue of lamellar phases. In that article a mode of “slip” between the two constituent elements (surfactants and water) was introduced. Lamellar phase hydrodynamics has been further discussed by Roux et al [10] [11].

Elasticity

The surfactant bilayers are arranged in a symmetric structure of smectic A. Taking z to describe the axis perpendicular to the layers, one layer deformation is described by $u(x, y)$, where u is the surface height compared to its original plane at point (x, y) . Since there are many layers we describe the layers deformation by a scalar field $u(x, y, z)$, where the x, y, z coordinates point to different segments of the lamellar phase.

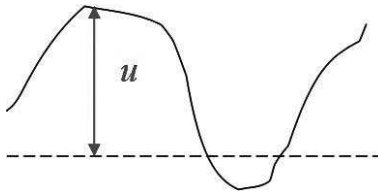


Figure 2: Layer displacement

A change in the elastic energy results from changes in u . The first derivatives $\frac{\partial u}{\partial x}, \frac{\partial u}{\partial y}$ could be created only from translation or rotation of the entire phase. Therefore, they cannot appear in the energy expression. From these symmetry considerations and Hooke’s law, small deviations from equilibrium cause the elastic energy to contain expressions quadratic in the layers deformations’ second derivatives.

Bending

The expression for a layer elastic energy does not depend on the coordinates one chooses. The differential geometry of a surface supply us with two such invariants: The Gauss curvature (\tilde{K}) and the mean curvature (H). An almost flat surface, (like the lamellar’s bilayers) may be described by $z = u(x, y)$ where the partial derivatives u_x, u_y satisfy $u_x, u_y \ll 1$.

These surface curvatures are [5]:

$$\tilde{K} \simeq u_{xx}u_{yy} - u_{xy}^2 \quad H \simeq u_{xx} + u_{yy} = \nabla_{\perp}^2 u$$

The expression for H is first order in u , while \tilde{K} is second order in u . Since ∂u is small $H \gg \tilde{K}$. For each membrane the bending energy will be a constant κ multiplying the mean curvature (H) squared. The lamellar phase is constructed from bilayers in a distance d from each other, therefore the constant of the bending elastic energy per unit length in \hat{z} is $K = \frac{\kappa}{d}$ (not to be confused with Gauss curvature \tilde{K} .)

Compressibility

The elastic compression energy emerging from the change in membranes' spacing will be described by $(\frac{\partial u}{\partial z})^2$. This expression includes only a first derivative, and is squared because of Hooke's law. The expression $(\frac{\partial u}{\partial z})^2$ is multiplied by a constant B , whose value is determined by the strength of the attraction and repulsion interactions between the layers. To conclude, the elastic energy per unit volume is:

$$\frac{1}{2}B \left(\frac{\partial u}{\partial z} \right)^2 + \frac{1}{2}K(\nabla_{\perp}^2 u)^2$$

The elastic force is given by $-\frac{\delta E}{\delta u} \hat{z}$:

$$\mathbf{F} = \left(B \frac{\partial^2 u}{\partial z^2} - K \nabla_{\perp}^2 (\nabla_{\perp}^2 u) \right) \hat{z}$$

This can be described by a divergence of an elastic tensor $\mathbf{F} = \nabla \cdot \sigma^{\text{el}}$, with:

$$\begin{cases} \sigma_{xx}^{\text{el}} = \sigma_{yy}^{\text{el}} = K \nabla_{\perp}^2 \frac{\partial u}{\partial z} & \sigma_{zz}^{\text{el}} = B \frac{\partial u}{\partial z} \\ \sigma_{xz}^{\text{el}} = -K \nabla_{\perp}^2 \frac{\partial u}{\partial x} & \sigma_{yz}^{\text{el}} = -K \nabla_{\perp}^2 \frac{\partial u}{\partial y} & \sigma_{xy}^{\text{el}} = 0 \end{cases} \quad (8)$$

Viscosity

Viscous forces appear when the velocity is not uniform. Resulting from symmetry considerations, the viscous stress tensor depends on the symmetric terms

$$v_{ij} = \frac{1}{2} \left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right)$$

Therefore, the most general expression for the viscous stress tensor σ' has the form

$$\sigma'_{\alpha\beta} = v_{\gamma\delta} \eta_{\alpha\beta\gamma\delta}$$

Unlike simple fluid, the viscosity in the lamellar phase is not isotropic. For any uniaxial phase the viscous tensor's terms involve five independent viscosities [4]. Martin et al [8] used the notation

$$\begin{aligned} \sigma'_{ij} = & 2\eta_2 v_{ij} + 2(\eta_3 - \eta_2)(v_{iz}\delta_{jz} + v_{jz}\delta_{iz}) + (\eta_4 - \eta_2)\delta_{ij}v_{kk} \\ & + (\eta_1 + \eta_2 - 4\eta_3 - 2\eta_5 + \eta_4)\delta_{iz}\delta_{jz}v_{zz} \\ & + (\eta_5 - \eta_4 + \eta_2)(\delta_{ij}v_{zz} + \delta_{iz}\delta_{jz}v_{kk}) \end{aligned}$$

For incompressible fluid $v_{kk} = 0$ and the viscous stress tensor takes the form:

$$\alpha, \beta \neq z \quad \left\{ \begin{array}{l} \sigma'_{zz} = (\eta_1 - \eta_5)v_{zz} \\ \sigma'_{\alpha z} = 2\eta_3 v_{\alpha z} \\ \sigma'_{\alpha\beta} = 2\eta_2 v_{\alpha\beta} + (\eta_5 - \eta_4 + \eta_2)\delta_{\alpha\beta}v_{zz} \end{array} \right.$$

A constant diagonal matrix can be subtracted from the viscous tensor and be included in the pressure. Subtracting $(\eta_5 - \eta_4 + \eta_2)v_{zz}$ from σ'_{xx} σ'_{yy} σ'_{zz} leads to Brochard and de-Gennes's expression for the incompressible lamellar phase viscous stress tensor [3]:

$$\alpha, \beta \neq z \quad \left\{ \begin{array}{l} \sigma'_{zz} = 2\eta_V \frac{\partial v_z}{\partial x_z} \\ \sigma'_{\alpha z} = \eta_M \left(\frac{\partial v_z}{\partial x_\alpha} + \frac{\partial v_\alpha}{\partial x_z} \right) \\ \sigma'_{\alpha\beta} = \eta_T \left(\frac{\partial v_\beta}{\partial x_\alpha} + \frac{\partial v_\alpha}{\partial x_\beta} \right) \end{array} \right. \quad (9)$$

This change of notation is given by $\eta_M = \eta_3$, $\eta_T = \eta_2$ and $2\eta_V = \eta_1 - \eta_2 + \eta_4 - 2\eta_5$. For lamellar phase η_M is approximately water viscosity and η_T, η_V are much larger. The latter notation for the viscous stress tensor is used in this study.

Permeation

The motion of the fluid relative to the layers' pattern is called permeation. This mechanism was suggested by Helfrich [12], and further developed by Martin et al [8].

The flow of matter through the layer structure is both due to lipid molecules hopping from one layer to the other and due to the solvent molecules flowing through the layers. For the lamellar phases which interest us both these processes are extremely weak, on account of the small solubility of water in the fat regions and vice versa. For this reason they are neglected when we develop the surface modes.

The permeation flow is induced by the elastic force in the direction normal to the layers as described in [8] and [9]. Taking the normal in the z direction, the permeation is given by:

$$\dot{u} - v_z = \lambda_p \left(B \frac{\partial^2 u}{\partial z^2} - K \nabla_{\perp}^2 \nabla_{\perp}^2 u \right)$$

The parameter λ_p is called the permeability parameter. It controls the strength of permeation in a given smectic liquid crystal phase.

The permeation processes is necessary when studying flows directed normal to the layers. The permeation would be taken into consideration in the preliminary model for myelin formation, suggested in Appendix D. If the effect of diffusion of lipid molecules is neglected (as assumed in the myelin formation model), the permeation occur only due to solvent's flow through the lipid layers.

Slip

The lamellar phase has two components: the surfactants, forming the layers, and the solvent (mainly water), occupying the space between the layers. Along any plane parallel to the layers there is only one component (either surfactants or water). This yields an extra collective degree of freedom, existing in lamellar phases, that exists neither in (thermotropic) smectic A nor in isotropic binary mixtures (Fig 3).

Surfactants' migration in the layers' plane relative to water can be done by slipping. The tangent force f_{\perp} , driving this process, emerges from a 2D pressure field ψ existing in the two dimensional fluid of surfactants at each bilayer,

$$\mathbf{f}_{\perp} = \nabla_{\perp} \psi$$

The force f_{\perp} acts on the surfactant fraction. If the 2D fluid forming the layer is taken as

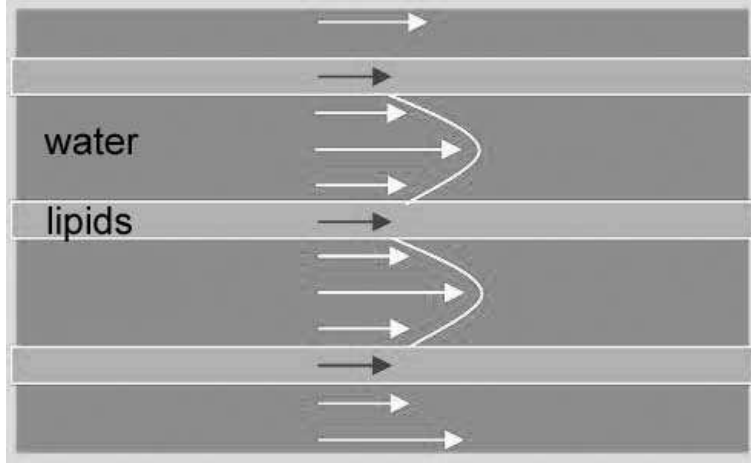


Figure 3: Slip

incompressible and containing no 2D viscous stresses, the induced slip flow is:

$$\mu \mathbf{f}_\perp = \mathbf{v}_{L\perp} - \mathbf{v}_\perp$$

\mathbf{v}_\perp and $\mathbf{v}_{L\perp}$ describes the average lamellar velocity parallel to the layers and the surfactants' velocity inside the layers respectively. The mobility is given by the slip coefficient μ .

Following [3], the slip coefficient can be estimated by considering a microscopic laminar flow of water relative to the bounding surfactant layers. Taking the z axis perpendicular to the layers and setting $z = 0$ at equal distances from the two layers, this flow has the form:

$$v_R = v_\perp \left(1 - \left(\frac{2z}{d} \right)^2 \right) - v_{L\perp} \quad (10)$$

Where v_R is the relative velocity, $v_{L\perp}$ is the lipid layers' velocity, v_\perp the maximal velocity of water, and d is the distance between the layers.

The driving force f_\perp of this relative flow is balanced by a viscous stress:

$$f_\perp = \eta_W \nabla^2 v_R \sim \frac{\eta_W}{d^2} v_R$$

Where we denote water viscosity by η_W . The slip coefficient deduced from the above equation is $\mu \sim \frac{d^2}{\eta_W}$. In a more detailed calculation (found in Ref. [3]), it is shown that if the layers' width is small compared to the distance between the layers $\mu = \frac{d^2}{12\eta_M}$

An extra equation is the mass conservation of the surfactants inside the layers (we ignore surfactants diffusion between layers). Considering each bilayer as a two dimensional incompressible fluid, gives the mass conservation equation:

$$\nabla_{\perp} \cdot \mathbf{v}_{L\perp} = 0$$

The contribution of the slip to the stress tensor is only in the diagonal terms σ_{xx} and σ_{yy} , both equal to ψ as expected from two dimensional pressure field. This pressure cannot induce forces on a surface parallel to the layers.

Summary of Lamellar Phase Hydrodynamics

We gather here the hydrodynamic equations relevant for lamellar phases, which are, except for the slip addition, relevant also for smectic A.

In Navier-Stokes equation: $\rho \frac{\partial \mathbf{v}}{\partial t} = -\nabla p + \nabla \cdot \sigma^{\text{el}} + \nabla \cdot \sigma'$ the non-linear term of the inertia is omitted, since this work refers to small perturbations on equilibrium state. The divergences of the elastic stress tensor Eq. 8 and the viscous stress tensor Eq. 9 are expanded in this subsection. We shall also assume incompressible fluid and no gravitation influence.

Mass conservation

$$0 = \nabla \cdot \mathbf{v} \quad (11)$$

Mass conservation for the surfactant fraction

$$0 = \nabla_{\perp} \cdot \mathbf{v}_{L\perp} \quad (12)$$

Navier-Stokes in x direction

$$\rho \frac{\partial v_x}{\partial t} = -\frac{\partial p}{\partial x} + \frac{\partial \psi}{\partial x} + 2\eta_T \frac{\partial^2 v_x}{\partial x^2} + \eta_T \left(\frac{\partial^2 v_y}{\partial x \partial y} + \frac{\partial^2 v_x}{\partial y^2} \right) + \eta_M \left(\frac{\partial^2 v_x}{\partial z^2} + \frac{\partial^2 v_z}{\partial x \partial z} \right) \quad (13)$$

Navier-Stokes in y direction

$$\rho \frac{\partial v_y}{\partial t} = -\frac{\partial p}{\partial y} + \frac{\partial \psi}{\partial y} + 2\eta_T \frac{\partial^2 v_y}{\partial y^2} + \eta_T \left(\frac{\partial^2 v_y}{\partial y^2} + \frac{\partial^2 v_x}{\partial x \partial y} \right) + \eta_M \left(\frac{\partial^2 v_y}{\partial z^2} + \frac{\partial^2 v_z}{\partial y \partial z} \right)$$

Navier-Stokes in z direction

$$\begin{aligned} \rho \frac{\partial v_z}{\partial t} = & -\frac{\partial p}{\partial z} + B \frac{\partial^2 u}{\partial z^2} - K \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) \left(\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} \right) \\ & + \eta_M \left(\frac{\partial^2 v_z}{\partial x^2} + \frac{\partial^2 v_z}{\partial y^2} + \frac{\partial^2 v_x}{\partial x \partial z} + \frac{\partial^2 v_y}{\partial y \partial z} \right) + 2\eta_V \frac{\partial^2 v_z}{\partial z^2} \end{aligned} \quad (14)$$

Permeation

$$\frac{\partial u}{\partial t} - v_z = \lambda_p \left(B \frac{\partial^2 u}{\partial z^2} - K \nabla_{\perp}^2 \nabla_{\perp}^2 u \right) \quad (15)$$

Slip

$$\mu \nabla_{\perp} \psi = \mathbf{v}_{L\perp} - \mathbf{v}_{\perp} \quad (16)$$

We did not include here phenomena of diffusion in each layer, and transfer of surfactant from one layer to the other.

2.5 Interfacial Boundary Conditions

In systems containing more than one phase, the hydrodynamics inside each phase can be described by its bulk hydrodynamic differential equations. At the interface the differential description is not valid anymore. In order to deduce the boundary conditions at the interface we need to go back to the integral form of the hydrodynamic equations.

We denote the interface sides by (+) and (-). The normal to the interface is chosen toward the (+) side. Let v_s be the interface velocity. Each phase is characterized by its density ρ and its stress tensor σ .

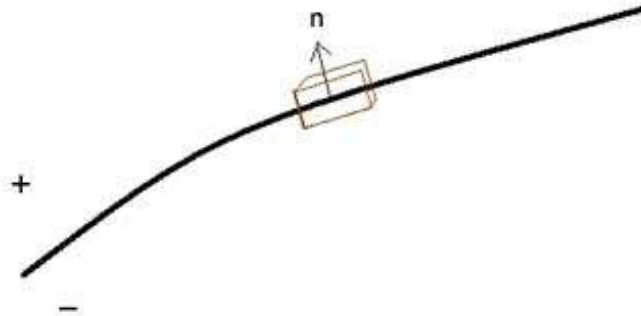


Figure 4: Interface

Imagine a small box whose bases are in different sides of the interface (Fig. 4). The base area is A and the box height is ϵ . We denote the volume inside the box in the (+) side

(respectively $(-)$ side) by V_+ (respectively V_-).

Boundary Condition due to Mass Conservation

The mass conservation equation in its integral form (Eq. 1), integrated over our box, has the form:

$$\iiint \frac{\partial \rho}{\partial t} dV = - \iint \rho \mathbf{v} \cdot \hat{\mathbf{n}} dA$$

Taking the box's height ϵ to zero eliminates the volume integral. This gives the boundary condition:

$$(\rho_+(\mathbf{v}_+ - \mathbf{v}_s) - \rho_-(\mathbf{v}_- - \mathbf{v}_s)) \cdot \hat{\mathbf{n}} = 0$$

For phases with equal density this condition reduces to: $(\mathbf{v}_+ - \mathbf{v}_-) \cdot \hat{\mathbf{n}} = 0$ meaning, the velocity normal to the interface is equal in both sides of the interface.

Boundary Condition due to Navier-Stokes

In Navier-Stokes equation (Eq. 3), the elements integrated on the volume are eliminated by taking ϵ to zero. At the interface, there is another force - the surface tension. The surface tension can be defined as the energy needed in order to add a unit area to the interface. Following [7], for approximately flat surface parallel to x - y plane the vertical force exerted by surface tension is: $\gamma(\frac{\partial^2 \zeta}{\partial x^2} + \frac{\partial^2 \zeta}{\partial y^2})$, where ζ is the interface's deviation from x - y plane and γ is the surface tension coefficient. Using Cauchy theorem (Eq. 4) we obtain the condition:

$$(\sigma_+ - \sigma_-) \hat{\mathbf{n}} = \gamma \left(\frac{\partial^2 \zeta}{\partial x^2} + \frac{\partial^2 \zeta}{\partial y^2} \right) \hat{\mathbf{n}}$$

This means that the stress tensor's elements σ_{xz} and σ_{yz} are equal in both sides of the interface, and the element σ_{zz} is discontinuous because of the surface tension.

3 Model

In this work we study the surface modes in a system of a semi infinite lamellar phase or smectic A phase having an interface with a semi infinite phase of gas or simple fluid. Let the interface be at the x - y plane, and the liquid crystal at $z < 0$. The liquid crystal's layers are parallel to the interface. In order to achieve the surface modes, we need to solve the bulk hydrodynamic equations of each phase (apart from the gas) and the boundary conditions equations.

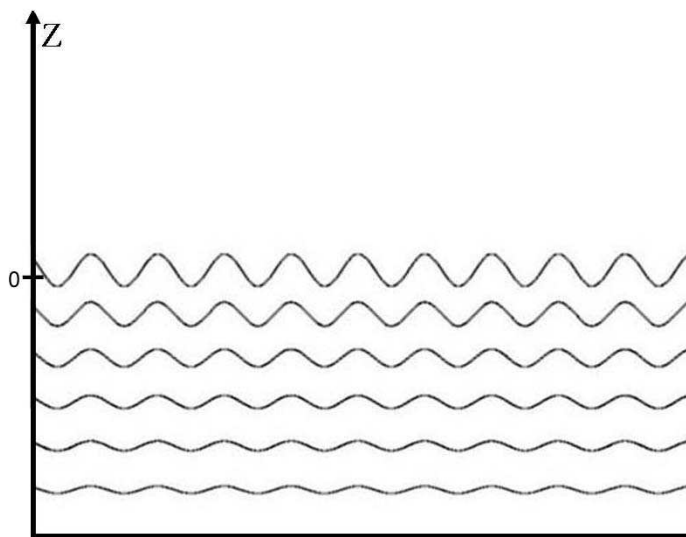


Figure 5: Surface modes

To a good approximation these phases (apart from the gas), could be taken as incompressible. The hydrodynamic equations describing small fluctuations on equilibrium of these phases are described in the background section. For lamellar phase these are equations 11-16, and for simple fluid these are equations 6 and 7. We shall consider a flow only in x and z directions, i.e., the system is uniform in the y direction. We denote the lamellar phase by "L", the simple fluid phase by "S" and the gas phase by "G". The boundary condition Eq. 2.5, applied to these systems, gives:

1. Continuity in the tangent forces (σ_{xz})

$$\sigma_{xz L} = \sigma_{xz S/G} \quad (17)$$

2. Discontinuity in the normal force (σ_{zz})

$$\sigma_{zz L} - \sigma_{zz S/G} - \gamma \frac{\partial^2 u}{\partial x^2} = 0 \quad (18)$$

where $\gamma \frac{\partial^2 u}{\partial x^2}$ is the force due to surface tension.

For lamellar phase -

$$\begin{aligned} \sigma_{xz} &= \eta_M \left(\frac{\partial v_x}{\partial z} + \frac{\partial v_z}{\partial x} \right) - K \frac{\partial^3 u}{\partial x^3} \\ \sigma_{zz} &= 2\eta_V \frac{\partial v_z}{\partial z} - p + B \frac{\partial u}{\partial z} \end{aligned} \quad (19)$$

For simple fluid -

$$\begin{aligned} \sigma_{xz} &= \eta \left(\frac{\partial v_x}{\partial z} + \frac{\partial v_z}{\partial x} \right) \\ \sigma_{zz} &= 2\eta \frac{\partial v_z}{\partial z} - p \end{aligned}$$

When the upper phase is a dilute gas, only its (assumed constant) pressure affects the lamellar phase by $\sigma_{zz G} = -p_{\text{gas}}$. The influence of the gas ability to compress and its viscosity are negligible in the dynamics description. In this case the boundary conditions are only on $\sigma_{xz L}$ and $\sigma_{zz L}$.

When the upper phase is a simple fluid, the mass conservation condition Eq. 2.5 is added to the above two boundary conditions. Assuming both phases densities are roughly equal, which is true for lamellar phase and its solvent, this condition yields a third demand:

3. Continuity in the normal velocity

$$v_z L = v_z S \quad (20)$$

In the case of lamellar phase and a simple fluid, additional boundary conditions are required. One is provided by tangent velocity continuity, arising from the two phases viscosity, since a jump in v_x will entail an infinite viscous stress. In cases where permeation is allowed through the interface, the pressure should be continuous as well. The additional boundary conditions are therefore:

4. Continuity in the tangent velocity

$$v_{xL} = v_{xS} \tag{21}$$

5. Pressure continuity

$$p_L = p_S$$

4 Dimensional Analysis

The surface modes analysis in this work is performed in two stages. In the first stage we do not include the slip property of the lamellar phase. Dropping the slip property reduces the hydrodynamic description to that of a single-component smectic A phase. (In order to examine the obtained modes we use, though, the lamellar's typical parameters.) To point out the applicability of the first stage for both Smectic A and Lamellar Phase we shall use a short notation - SA/LP .

In the second stage we examine the influence of the slip property on the surface modes. This analysis is relevant only for two-component smectic A, i.e., a lyotropic lamellar phase. In each stage we examine the case when $z > 0$ is occupied by gas, and the case when $z > 0$ is occupied by simple fluid.

Contrary to Fedorov et al [1], in the present study we focus on surface modes that are not influenced by inertia, i.e., on low Reynolds numbers. In order to simplify the equations we have omitted the extremely weak process of permeation.

The parameters that characterize the surface dynamics of SA/LP and gas are B , K , γ , η_T , η_V , η_M , ω and q . ω describes the time dependence, q is the wave number. From these parameters three dimensionless expressions involving ω can be established, corresponding to the three restoring forces in the system:

$$\text{De}_1^{-1} = \frac{-iB}{\eta_M\omega} \quad \text{De}_2^{-1} = \frac{-iKq^2}{\eta_M\omega} \quad \text{Ca} = \frac{-i\gamma|q|}{\eta_M\omega} \quad (22)$$

De_1 and De_2 are Deborah numbers, associated with the visco elastic properties. Ca is a capillary number, associated with the surface tension. Since η_T and η_V appear in a symmetric form in the equation for ω (further discussed), the dimensionless parameter for the SA/LP viscosities is written as $\Theta = \frac{2(\eta_T + \eta_V)}{\eta_M}$. In the case of SA/LP and simple fluid, another dimensionless parameter appears due to the simple fluid viscosity $s = \frac{2\eta}{\eta_M}$ where η is the simple fluid viscosity. While η_T and η_V are much larger than the solvent (water) viscosity, η_M is comparable to it. Thus, Θ is always a large number ($10^2 - 10^3$), and s is typically of order 1 (unless the contact simple fluid is a very viscous one). Variations on the viscosity term in the dimensionless parameters Eq. 22 could be obtain by Θ and s ; the choice of η_M in

Eq. 22 is arbitrary. The inertia dimensionless parameter is the Reynolds number $\text{Re} = \frac{i\omega\rho}{\eta_M q^2}$.

Since small fluctuations on the equilibrium state should decay in time, we expect to find ω to be a negative imaginary number. For such ω the above dimensionless expressions are positive. (Actually, ω sometimes have also a real part, meaning damped oscillations in time. Real parts are found beyond first order approximations.)

Analysis of the dimensionless parameters lead to three limiting cases for the decay rates, when one restoring force predominates:

$$\omega_B = \frac{-iB}{\eta'} \quad \omega_K = \frac{-iKq^2}{\eta''} \quad \omega_\gamma = \frac{-i\gamma|q|}{\eta'''} \quad (23)$$

The specific viscosity parameters entering these expressions and their validity as surface modes will be determined.

The slip dimensionless parameter is $m = (\eta_M \mu q^2)^{-1}$. This parameter competes with the lamellar's anisotropic viscosity parameter Θ (as shown below). When the slip property is considered the temporal decay rates change significantly.

In order to compare the liquid crystal's surface modes to simple fluid surface modes, we wish to discuss also the latter modes. Neglecting inertia, we get from the simple fluid bulk equations (Eq. 6) and the boundary conditions (Eq. 2.5, 2.5) one surface mode. In the case of simple fluid and gas, the frequency is

$$\omega = \frac{-i\gamma|q|}{2\eta} \quad (24)$$

In the case of two simple fluid phases, where the viscosities are η_1 and η_2 , the surface mode decay is given by

$$\omega = \frac{-i\gamma|q|}{2(\eta_1 + \eta_2)} \quad (25)$$

This mode for simple fluid is analogous to ω_γ in Eq. 23 for SA/LP .

The spatial decay in the z axis for simple fluid is equal to the wave number q , (dependence of $\exp(qz)$). This is because wave length is the single length parameter for the simple fluid system. In SA/LP , there are other three typical lengths: $\sqrt{K/B}$, γ/B , and K/γ , variations on these lengths can be achieved by the viscosity dimensionless parameters. For lamellar phases there is another length parameter given by the slip property which is $\sqrt{\eta_M \mu}$.

The range of wave lengths we refer to is restricted from below by the continuous description of hydrodynamics to approximately 10^{-8} meter. The area where inertia is no longer negligible will give the upper bound.

Experimental data on characteristics of lamellar phases and on simple fluids provide approximations of their parameters [4], [10], [11], [13], [14]:

$$\begin{aligned}
 B &\sim 10^3 - 10^7 [Pa], & K &\sim 10^{-13} [Pa/m^2], & \rho &\sim 10^3 [Kg/m^3] \\
 \eta_M &\sim 10^{-3}, & \eta_T &\sim 10^{-1}, & \eta_V &\sim 10^{-1} [Kg/(msec)] \\
 \gamma &< 10^{-3} - 10^{-1} [Pa/m]
 \end{aligned}
 \tag{26}$$

From the typical values in Eq. 26, and the minimum values of the wave length as required for the hydrodynamic treatment, we can always assume $De_1^{-1} \gg De_2^{-1}$. We also use $\Theta \gg 1$ in our approximations.

5 Hydrodynamic Variables and Spatial Decay

In this section, the connections between the hydrodynamic variables, and the relation between the spatial and temporal decays are deduced. An equation for the decay rate is obtained for the interfaces between SA/LP and simple fluid and between SA/LP and gas. The slip property is omitted at this stage, we consider it later on in section 8.

5.1 The Lamellar Bulk Dynamics

The surface modes are small fluctuations on the equilibrium state. Since \mathbf{v} , u and ∇p in equilibrium are zero, the hydrodynamic variables are henceforth small perturbations on the equilibrium state. We are interested in the leading (linear) response to these perturbations.

We explore the effect of a small arbitrary fluctuation by performing a Fourier Transform (FT) on the hydrodynamic variables in time and space:

$$(v_z, v_x, u, p) \rightarrow \int dx dt (v_z, v_x, u, p) e^{iqx} e^{-i\omega t}$$

The transform on x coordinate is allowed due to the continuity in the hydrodynamic description along the x direction.

The constant pressure at equilibrium is equal in both phases and therefore can be omitted. The set of equations (Eq. 12- 15) gives the relations between the four hydrodynamic functions $v_z(z)$, $v_x(z)$, $u(z)$ and $p(z)$. Isolating $v_z(z)$ yields a presentation of the rest of the functions:

$$\begin{aligned} u(z) &= -i\omega v_z(z) \\ v_x(z) &= \frac{i}{q} v_z(z) \\ p(z) &= \frac{1}{q} \left(\left(\frac{i\omega\rho}{q} - 2\eta_T q + \eta_M q \right) \frac{\partial v_z(z)}{\partial z} + \frac{\eta_M}{q} \frac{\partial^3 v_z(z)}{\partial z^3} \right) \end{aligned} \quad (27)$$

The resulting equation for v_z is:

$$\begin{aligned} 0 &= v_z(z) \left(\frac{iKq^4}{\omega} + q^2 \eta_M - i\omega\rho \right) \\ &+ \frac{\partial^2 v_z(z)}{\partial z^2} \left(\frac{i\omega\rho}{q^2} - \frac{iB}{\omega} - 2\eta_T - 2\eta_V + 2\eta_M \right) + \frac{\partial^4 v_z(z)}{\partial z^4} \frac{\eta_M}{q^2} \end{aligned} \quad (28)$$

This is a fourth order ODE containing only even order derivatives (0, 2, 4). We try a solution of the form $v_z(z) = A \exp(\alpha z)$ and get a second order polynomial for α^2 . This polynomial has four solutions for α .

Surface modes decay as we go away from the interface. Since in our description the SA/LP is in the area of negative z , the possible spatial decay (α) must have a positive real part. We are left with two possible decays:

$$\alpha_{1,2} = |q| \sqrt{\frac{1}{2}\Theta - 1 - \frac{1}{2}\text{De}_1^{-1} - \frac{1}{2}\text{Re} \pm \sqrt{\Upsilon}} \quad (29)$$

where $\Upsilon = (\frac{1}{2}\Theta - 1 - \frac{1}{2}\text{De}_1^{-1} - \frac{1}{2}\text{Re})^2 - (1 - \text{De}_2^{-1} - \text{Re})$.

Therefore, we have $v_z(z) = C_1 \exp(\alpha_1 z) + C_2 \exp(\alpha_2 z)$. C_1 and C_2 set the amplitude of each spatial decay. They are determined by the boundary conditions at the interface.² The boundary conditions determine also the relations between the spatial decay coefficients (α_1, α_2) and the expression for the temporal decay (ω) i.e., the dispersion relation. In the next sub-sections the equation for the temporal decay is derived.

5.2 Equations for Interfaces Between SA/LP and Gas

The gas phase dynamics is not relevant to describe the SA/LP surface modes. The boundary conditions (Eq. 17, Eq. 18) at the interface ($z = 0$) are:

$$\begin{aligned} \sigma_{xz} L = 0 &\Rightarrow \sum_{j=1}^2 \left(i\eta_M \left(q + \frac{\alpha_j^2}{q} \right) - \frac{Kq^3}{\omega} \right) C_j = 0 \\ \sigma_{zz} L - \gamma \frac{\partial^2 u}{\partial z^2} = 0 &\Rightarrow \sum_{j=1}^2 \left(\alpha_j (2\eta_V + 2\eta_T - \eta_M + \frac{iB}{\omega}) \right. \\ &\quad \left. - \frac{\eta_M}{q^2} \alpha_j^3 + \frac{i\gamma q^2}{\omega} \right) C_j = 0 \end{aligned} \quad (30)$$

The expressions of the stress tensor elements used here are written in Eq. 19.

This pair of equations can be written as a matrix multiplied by the coefficient vector (C_1, C_2) . A trivial solution would be when the coefficients are zero and therefore there is

²Rigorously, the possibility of only one spatial decay length ($v_z(z) = (a+bz) \exp(\alpha z)$) should be considered as well. This option determines the spatial and temporal decays already from the bulk equations and, in general (similar to the case of simple fluids), will not satisfy the boundary conditions, as shown in Appendix A.

no fluctuation at all. We demand the vanishing of the matrix determinant in order to get a non-trivial solution. This determines the dispersion relation $\omega(q)$. The resulting equation expressed in dimensionless parameters is:

$$\text{Ca} \frac{\alpha_1 + \alpha_2}{|q|} + \text{Re} - \sqrt{1 - \text{De}_2^{-1} - \text{Re}(\Theta - \text{De}_1^{-1} - \text{De}_2^{-1} - \text{Re})} = 0 \quad (31)$$

where $\frac{\alpha_{1,2}}{|q|}$ are presented in Eq. 29.

5.3 Equations for Interfaces Between SA/LP and Simple Fluid

The simple fluid hydrodynamics is given by Eq. 6, Eq. 7. Including inertia there are two spatial decays for the simple fluid phase. The hydrodynamic functions' spatial decay has the form:

$$A \exp(-|q|z) + B \exp\left(-\sqrt{q^2 - \frac{i\omega\rho}{\eta}}z\right)$$

If the inertia is not included, there is only one spatial decay. In this case the hydrodynamic functions spatial dependence has the form:

$$(A + Bz) \exp(-|q|z) \quad (32)$$

Since the solution for $\rho \neq 0$ does not converge continuously to the one for $\rho = 0$ as $\rho \rightarrow 0$, we use Eq. 32 (i.e., $\text{Re} = 0$) from the beginnings. Therefore, in the case of SA/LP and simple fluid system, we shall ignore inertia from the beginning.

The connections between the simple fluid hydrodynamic variables are:

$$v_x(z) = \frac{i}{q} \frac{\partial v_z(z)}{\partial z}$$

$$p(z) = -\eta \frac{\partial v_z(z)}{\partial z} + \frac{\eta}{q^2} \frac{\partial^3 v_z(z)}{\partial z^3}$$

There are two additional boundary conditions in this case: the continuity of the normal and the tangent components of the velocity (Eq. 20 and Eq. 21).

The determinant giving $\omega(q)$ from the matrix description of the boundary conditions is:

$$s^2 - \text{Cas} - \text{Ca} \frac{\alpha_1 + \alpha_2}{|q|} + s \frac{\alpha_1 + \alpha_2}{|q|} + \sqrt{1 - \text{De}_2^{-1}} (-\text{De}_1^{-1} - \text{De}_2^{-1} + s \frac{\alpha_1 + \alpha_2}{|q|} + \Theta) = 0 \quad (33)$$

where $\frac{\alpha_{1,2}}{|q|}$ are the SA/LP spatial decays, derived when the inertia is neglected:

$$\alpha_{1,2} = |q| \sqrt{\frac{1}{2}\Theta - 1 - \frac{1}{2}\text{De}_1^{-1} \pm \sqrt{\left(\frac{1}{2}\Theta - 1 - \frac{1}{2}\text{De}_1^{-1}\right)^2 - (1 - \text{De}_2^{-1})}} \quad (34)$$

In the next sections the solution of these determinant equations will be developed.

6 Decay Rate of Surface Modes Between SA/LP and Gas

Eq. 31 can be transformed to a six degree polynomial equation. Unfortunately, the prominent 19th century mathematician Galois, has proved at the night before the duel where he found his death, that a general formula for the roots of a polynomial, expressed by the polynomial's coefficients can be found only if the degree of the polynomial is at most four. Therefore, our equation for the temporal decay cannot, actually, be solved analytically. Eq. 31 could be solved numerically. We prefer to study it by examining limiting cases, in order to achieve understanding of the processes governing the system.

We shall develop Eq. 31 asymptotically in three limiting cases, detailed in Eq. 23:

- When the decay is governed by the membranes' ability to bend. This rate has the form of ω_K .
- When the decay is governed by the membranes' resistance to compression. This rate has the form of ω_B .
- When the decay is governed by the surface tension. This rate has the form of ω_γ .

Corrections to each limiting case are found by second order approximations in the non governing restoring mechanisms.

Since we do not investigate modes that depend on inertia, we look at Eq. 31 without the inertial part Re . Neglecting inertia leads to the equation:

$$\text{Ca} \frac{\alpha_1 + \alpha_2}{|q|} - \sqrt{1 - \text{De}_2^{-1}(\Theta - \text{De}_1^{-1} - \text{De}_2^{-1})} = 0$$

$$\frac{\alpha_{1,2}}{|q|} = \sqrt{\frac{1}{2}\Theta - 1 - \frac{1}{2}\text{De}_1^{-1} \pm \sqrt{(\frac{1}{2}\Theta - 1 - \frac{1}{2}\text{De}_1^{-1})^2 - (1 - \text{De}_2^{-1})}}$$
(35)

α_1 corresponds to the plus sign in the expression inside the root, and α_2 to the minus sign.

When the surface tension is neglected, $\text{Ca} \rightarrow 0$, Eq. 35 is reduced to the form:

$$\sqrt{1 - \text{De}_2^{-1}(\Theta - \text{De}_1^{-1})} = 0$$
(36)

The element De_2^{-1} has been neglected here compared to De_1^{-1} .

6.1 Bending-Dominated Rate

First order approximation

The solution emerging from equating De_2^{-1} with 1 in Eq. 36 is:

$$\omega = \omega_K = -\frac{iKq^2}{\eta_M} \quad (37)$$

The spatial decays for this mode are found from Eq. 35 to be:

$$\alpha_1 = |q| \sqrt{\Theta - 2 - \frac{B}{Kq^2}}, \quad \alpha_2 = 0 \quad (38)$$

For this mode to be a surface mode both α_1 and α_2 should have a positive real part. The only way this mode will be a surface mode, then, is if the amplitude coefficient for α_2 spatial decay (C_2) is zero. Placing ω_K in the tangent force boundary condition (Eq. 30) gives the relation:

$$C_1 \alpha_1^2 = -C_2 \alpha_2^2 \quad (39)$$

Where C_1 and C_2 are the coefficients for the spatial decays α_1 and α_2 respectively. From Eq. 38 and Eq. 39 the coefficient C_1 is zero. Therefore, there is no spatial decay in this mode. The layers in this description have a sinusoidal deformation, uniform in the z direction:

$$u \propto e^{iqx} e^{-\frac{\kappa q^2}{\eta_M} t}$$

This mode is not a surface mode. It is a pure undulation bulk mode, that was found by Martin et al [8] and Brochard et al [3]. A derivation of this bulk mode is presented in appendix B.

Bending-Dominated mode with Correction due to Surface Tension

Second order approximation to the bulk undulation mode, which includes surface tension, leads to positive real spatial decays, i.e. to surface modes.

Under the approximation $\text{De}_1^{-1} \ll \Theta$ the spatial decay part is approximately:

$$\frac{\alpha_1 + \alpha_2}{|q|} \simeq \frac{\alpha_1}{|q|} \simeq \sqrt{\Theta}$$

Under the conditions:

$$\text{Ca}\sqrt{\Theta} \gg \max\{\text{Re}, \text{De}_1^{-1}\}, \quad \text{De}_1^{-1} \ll \Theta, \quad \text{Re} \ll 1 \quad (40)$$

The determinant takes the form:

$$\text{Ca}\sqrt{\Theta} - \sqrt{1 - \text{De}_2^{-1}\Theta} = 0 \quad (41)$$

Solving perturbatively in small Ca we reach the undulation mode with its correction due to surface tension:

$$\omega = \omega_{K,(\gamma)} = -\frac{iKq^2}{\eta_M} - \frac{i\gamma^2}{2K(\eta_T + \eta_V)} \quad (42)$$

In this approximation $\alpha_2 \simeq |q|\sqrt{(1 - \text{De}_2^{-1})/\sqrt{\Theta}}$ is a positive real expression, because $\text{De}_2^{-1} < 1$. This is, therefore, a surface mode because both spatial decays are positive.

For this to be indeed a correction to the pure undulation mode the element added to the temporal decay must be less significant than the temporal decay characterizing the undulation mode. This yields a very limiting condition:

$$|q| \gg \frac{\gamma}{K\sqrt{\Theta}} \quad (43)$$

The latter condition and the conditions from Eq. 40 confine $|q|$ to:

$$\max\left\{\sqrt{\frac{B}{K\Theta}}, \frac{B}{\gamma\sqrt{\Theta}}, \frac{\gamma}{K\sqrt{\Theta}}\right\} \ll |q| \ll \frac{\gamma\sqrt{\Theta}\eta_M^2}{K^2\rho} \quad (44)$$

Therefore, this mode could be present in systems in which:

$$\frac{\rho K}{\eta_M^2} \ll 1, \quad \max\left\{\sqrt{\frac{B}{K\Theta}}, \frac{B}{\gamma\sqrt{\Theta}}, \frac{\gamma}{K\sqrt{\Theta}}\right\} \ll \frac{\gamma\sqrt{\Theta}\eta_M^2}{K^2\rho}$$

This is a diffusive surface mode $\omega \sim q^2$. The spatial decays are $\alpha_1 \simeq |q|\sqrt{\Theta}$, $\alpha_2 \simeq \frac{\gamma}{K\Theta^{3/4}}$, where α_1 is larger than α_2 due to Eq. 43. The slow spatial decay α_2 is more dominant than α_1 as can be seen from Eq. 39, $|C_2| \gg |C_1|$. For typical parameters of smectic A and lamellar phase, the above conditions on $|q|$ and the large spatial decay given by α_2 make this mode unphysical.

6.2 Compression-Dominated Rate

First order approximation

The second solution to Eq. 36 is given by equating De_1^{-1} with Θ . This temporal decay is governed by compression.

$$\begin{aligned}\omega = \omega_B &= -\frac{iB}{2(\eta_T + \eta_V)} \\ \alpha_{1,2} &= |q| \sqrt{-1 \pm \sqrt{\text{De}_2^{-1}}}\end{aligned}\tag{45}$$

The boundary condition on tangent force at the interface (Eq. 30) gives the relation:

$$C_1 \left(\sqrt{\frac{B}{Kq^2}} + \sqrt{\Theta} \right) = C_2 \left(\sqrt{\frac{B}{Kq^2}} - \sqrt{\Theta} \right)$$

α_2 is imaginary and its amplitude does not vanish. Therefore, this is not a surface mode. This is a bulk compression mode discussed in Appendix C

Compression and Bending Dominated Rate

Under the approximation of small Ca , a surface mode is achieved by considering De_2^{-1} as non-negligible compared to De_1^{-1} . This approximation of Eq. 35, gives a solution in which:

$$\Theta - \text{De}_1^{-1} - \text{De}_2^{-1} = 0\tag{46}$$

The solution is:

$$\omega = \omega_{B,K} = -\frac{i(B + Kq^2)}{2(\eta_T + \eta_V)}\tag{47}$$

The spatial decays in this mode, given by Eq. 35, are:

$$\alpha_1 = |q| \sqrt{\text{De}_2^{-1} - 1} = |q| \sqrt{\frac{Kq^2\Theta}{B + Kq^2} - 1} \quad \alpha_2 = i|q|$$

Placing α_1 , α_2 and $\omega_{B,K}$ in the tangent force boundary condition Eq. 17, gives $C_2 = 0$. Thus, the spatial behavior corresponds to α_2 has a zero amplitude. Therefore, the spatial decay is given only by α_1 .

The conditions for $\omega_{B,K}$ to belong to a surface mode (simplified by $\text{De}_1^{-1} \gg \text{De}_2^{-1}$) are:

- $\text{Ca}_{|q|}^{\alpha_1} \ll \text{De}_2^{-1} |\sqrt{1 - \text{De}_2^{-1}}|$, for the current approximation to be valid.
- $\text{De}_2^{-1} > 1$, so that $\text{Re}(\alpha_1) > 0$.
- $\text{Re} \ll \text{De}_2^{-1}$ for the inertia to be negligible. This condition contains the requirement $\text{Re} \ll 1$ because, as a result of the previous condition, $\text{De}_2^{-1} > 1$.

These three conditions, and the demand $\text{De}_1^{-1} \gg \text{De}_2^{-1}$, give the following conditions on $|q|$:

$$|q| \gg \max\left\{\frac{\gamma}{K}, \sqrt{\frac{\rho B}{2\eta_M(\eta_T + \eta_V)}}\right\}, \quad |q| > \sqrt{\frac{B}{K\Theta}} \quad (48)$$

The above conditions confine the relevance of the solution to extremely low surface tension and small wave lengths, which are on the verge allowed in the hydrodynamic description.

6.3 Surface Tension Dominated Rate

First Order Approximation

In order to study the cases when the surface tension has the dominant role in determining the temporal decay, we shall keep the term $\text{Ca}_{|q|}^{\alpha_1 + \alpha_2}$ in the determinant equation (Eq. 31). Different approximations to Eq. 31 are obtained from the different asymptotic relations among the rest of the parameters: De_1^{-1} , De_2^{-1} , Θ and 1. The number of these approximations is reduced by the conditions $\text{De}_1^{-1} \gg \text{De}_2^{-1}$ and $\Theta \gg 1$, which can be assumed to hold always.

The relevant limits and their resulting first order determinant equations are:

1. $\text{De}_1^{-1} \ll \Theta$, $\text{De}_2^{-1} \ll 1$ \Rightarrow $\text{Ca}\sqrt{\Theta} - \Theta = 0$
2. $\text{De}_1^{-1} \gg \Theta$, $\text{De}_2^{-1} \ll 1$ \Rightarrow $\text{Ca}\sqrt{-\text{De}_1^{-1}} + \text{De}_1^{-1} = 0$
3. $\text{De}_1^{-1} \ll \Theta$, $\text{De}_2^{-1} \gg 1$ \Rightarrow $\text{Ca}\sqrt{\Theta} - \sqrt{-\text{De}_2^{-1}}\Theta = 0$
4. $\text{De}_1^{-1} \gg \Theta$, $\text{De}_2^{-1} \gg 1$ \Rightarrow $\text{Ca}\sqrt{-\text{De}_1^{-1}} - \sqrt{-\text{De}_2^{-1}}\text{De}_1^{-1} = 0$

For the last three cases there is no solution for ω . In case 4, ω cancels out. The approximate determinant equations, in cases 2 and 3 have no solution because one term in each equation is real and the other is imaginary. Expanding the approximate determinant equation of these

three cases further would not give a solution for ω either, since the additional terms in the expansion must be smaller than the leading ones.

The first case ($\text{De}_1^{-1} \ll \Theta$, $\text{De}_2^{-1} \ll 1$) is solvable. It gives a decay rate of:

$$\omega = \omega_\gamma = -\frac{i\gamma|q|}{\sqrt{2\eta_M(\eta_T + \eta_V)}} \quad (49)$$

This decay rate coincides with the decay rate for isotropic simple fluid Eq. 24 by taking $\eta_T = \eta_V = \eta_M = \eta$.

We obtain the amplitude (C_1 and C_2) ratio for the two spatial decays (α_1 and α_2) using the boundary condition of the tangent force continuity (Eq. 17). Under the approximation $\text{Ca} \gg \sqrt{\Theta}\text{De}_2^{-1}$, we have used in this case, the relation between the spatial decays' amplitudes given by Eq. 30 is:

$$(\alpha_2^2 + q^2)C_2 = -(\alpha_1^2 + q^2)C_1 \quad (50)$$

In light of the assumption $\text{De}_1^{-1} \ll \Theta$, $\text{De}_2^{-1} \ll 1$ and the anisotropic viscosity parameter $\Theta \simeq 10^2$, the spatial decays Eq. 35 obey $\alpha_1 \simeq \sqrt{\Theta}|q| > |q|$ and $\alpha_1 \gg \alpha_2$. The relation in Eq. 50 yields that the slow spatial decay α_2 has a larger amplitude ($|C_2| \gg |C_1|$). In order to find α_2 we shall expand the inner root in the spatial decays expression Eq. 35 in large Θ , (considering $\text{De}_2^{-1} \ll 1$ and $\text{De}_1^{-1} \ll \Theta$):

$$\sqrt{\left(\frac{1}{2}\Theta - 1 - \frac{1}{2}\text{De}_1^{-1}\right)^2 - (1 - \text{De}_2^{-1})} \simeq \frac{1}{2}\Theta - 1 - \frac{1}{2}\text{De}_1^{-1} - \frac{1}{\Theta}$$

This gives the second spatial decay $\alpha_2 \simeq \frac{1}{\sqrt{\Theta}}|q| < |q|$.

Since the two spatial decays are positive, this is a surface mode. This surface mode has $\omega \sim q$. The spatial decays are proportional to the wave number $\alpha_1, \alpha_2 \sim |q|$, having opposite phases, $\text{sign}(C_1) = -\text{sign}(C_2)$.

For this mode to be valid, we need to demand that all the terms we neglect in Eq. 31, are much smaller than the two left in this approximation. The validity region for this solution is therefore determined by:

$$\Theta \gg \text{De}_1^{-1} \quad \text{De}_2^{-1} \ll 1 \quad \text{Re} \ll 1$$

The conditions on $|q|$ given by these demands are:

$$\frac{B}{\gamma\sqrt{\Theta}} \ll |q| \ll \frac{\gamma}{K\sqrt{\Theta}} \quad |q| \gg \frac{\gamma\rho}{\eta_M^2\sqrt{\Theta}}$$

For this q range to be non zero the upper limit should be higher than the lower limit of q .

This gives two conditions on the system:

$$\gamma \gg \sqrt{BK}, \quad \frac{\rho K}{\eta_M^2} \ll 1$$

The allowed q range is very high, typically $|q| \gtrsim 10^5$, and the typical allowed values for ω are cotispondingly large, $\omega > \sim 10^6 \text{sec}^{-1}$ (fast decay). The typical spatial decays are on the order of $1\mu\text{m}$ to 1nm .

Elasticity Corrections for the Surface Tension-Dominated Rate

Expanding Eq. 31 to higher orders in the elastic parameters, keeping Ca and Θ as the most dominant parameters, gives the elastic correction to the surface tension dominated mode.

Correction for ω_γ due to bending gives the decay

$$\omega = \omega_{\gamma,(K)} = -\frac{i\gamma|q|}{\sqrt{2\eta_M(\eta_T + \eta_V)}} - \frac{iKq^2}{2\eta_M} \quad (51)$$

Correction for ω_γ due to compression gives the decay

$$\omega = \omega_{\gamma,(B)} = -\frac{i\gamma|q|}{\sqrt{2\eta_M(\eta_T + \eta_V)}} - \frac{iB}{2(\eta_V + \eta_T)} \quad (52)$$

Summary

A bending dominated, diffusive ($\omega \sim q^2$) surface mode is achieved by adding a correction to the pure undulation bulk mode due to surface tension (subsection 6.1). However, this is found not to be a physical surface mode.

The compression and bending dominated mode (subsection 6.2) is a fast-decaying mode governed by compression in which $\omega \sim |q|^0$, with a diffusive ($\sim q^2$) correction due to bending. It has one spatial decay. For large bending modulus the spatial decay is proportional to the wave number squared $\alpha \sim q^2\sqrt{K/B}$.

Typical lamellar phase parameters limit the compression and bending dominated mode to a region of high wave numbers ($|q| \gtrsim 10^7 \text{m}^{-1}$). The validity of the continuous hydrodynamic

description is thus sensitive to the parameters' values. Therefore, $\omega_{B,K}$ may become to a relevant surface mode only in systems with extremely low surface tension, high bending modulus, low compression modulus and high viscosity, where the exact conditions are given in Eq. 48. Only under these restrictive conditions is it valid for physically small wave lengths.

The viscosity anisotropy brings the elasticity dominated modes closer to the valid wave length region.

The surface tension dominated relaxation (subsection 6.3) is an “acoustic” ($\omega \sim q$) surface mode, with spatial decays that are proportional to the wave number $\alpha_1, \alpha_2 \sim |q|$. It is similar to the surface tension dominated mode for simple fluids Eq. 24. Its decay rate converge to Eq. 24 when the viscosity is made isotropic, and so do the spatial decays. This mode is valid for large wave numbers, typically $|q| \gtrsim 10^5 \text{m}^{-1}$. In this mode also, the viscosity anisotropy brings the relevant wave length regime into the region of hydrodynamics validity. From substituting the parameters for a lamellar phase in the conditions on $|q|$ we find that the surface tension dominated mode may be valid for $|q|$ larger than 10^5m^{-1} , and even this is true only for the lowest available B .

We present a summarize of these results in Table 1.

On the whole, our results in this chapter show, in agreement with Fedorov et al [1], that low-Reynolds-number surface relaxation modes in smectics are restricted to very narrow range of high $|q|$ and ω . Hence, surface relaxation in these systems is expected to actually be governed by inertia [1]. As we shall show in section 8, adding the slip property, that is relevant for the lamellar phase, will reveal modes that are not dominated by inertia.

	surface-tension-dominated relaxation
decay rate	$\omega_\gamma = -\frac{i\gamma q }{\sqrt{2\eta_M(\eta_T+\eta_V)}}$
spatial decays	$ q \sqrt{\Theta}, \quad \frac{ q }{\sqrt{\Theta}}$
$ q $ range	$\max\{\frac{B}{\gamma\sqrt{\Theta}}, \frac{\gamma\rho}{\eta_M^2\sqrt{\Theta}}\} \ll q \ll \frac{\gamma}{K\sqrt{\Theta}}$
system conditions	$\gamma \gg \sqrt{BK}$
remarks	relevant for large wave numbers only
	compression-dominated relaxation
decay rate	$\omega_{B,K} = -\frac{i(B+Kq^2)}{2(\eta_T+\eta_V)}$
spatial decay	$ q \sqrt{\frac{Kq^2\Theta}{B+Kq^2} - 1}$
$ q $ range	$ q \gg \max\{\frac{\gamma}{K}, \sqrt{\frac{\rho B}{2\eta_M(\eta_T+\eta_V)}}\}, \quad q > \sqrt{\frac{B}{K\Theta}}$
system conditions	-
remarks	practically irrelevant due to the restrictions on $ q $
	bending-dominated relaxation
decay rate	$\omega_{K(\gamma)} = -\frac{iKq^2}{\eta_M} - \frac{i\gamma^2}{2K(\eta_T+\eta_V)}$
spatial decays	$ q \sqrt{\Theta}, \quad \frac{\gamma}{K\Theta^{3/4}}$
$ q $ range	$\max\{\sqrt{\frac{B}{K\Theta}}, \frac{B}{\gamma\sqrt{\Theta}}, \frac{\gamma}{K\sqrt{\Theta}}\} \ll q \ll \frac{\gamma\sqrt{\Theta}\eta_M^2}{K^2\rho}$
system conditions	-
remarks	not a physical surface mode

Table 1: Results for smectic A

7 Decay Rate of Surface Modes Between SA/LP and Simple Fluid

The determinant equation for a system of SA/LP and simple fluid is given in Eq. 33:

$$s^2 - Cas - Ca \frac{\alpha_1 + \alpha_2}{|q|} + s \frac{\alpha_1 + \alpha_2}{|q|} + \sqrt{1 - De_2^{-1}(-De_1^{-1} - De_2^{-1} + s \frac{\alpha_1 + \alpha_2}{|q|} + \Theta)} = 0 \quad (33)$$

For a simple fluid with viscosity similar to the lamellar's solvent viscosity ($\eta \simeq \eta_M$), s is of order one. Assuming $s \equiv \frac{2\eta}{\eta_M} \simeq 1$ allows all modes found for SA/LP and gas, in essentially the same conditions. We shall repeat these modes briefly with their detailed approximation assumptions.

Correction due to the simple fluid viscosity are found for the surface tension dominated mode. For highly viscous simple fluids, the surface mode resembles the one created at the interface between two simple fluid phases, Eq. 25. In this mode the effective viscosity parameter appears as $\sqrt{2\eta_M(\eta_T + \eta_V)}$.

Unless mentioned otherwise, we shall assume $\eta \simeq \eta_M$, as is usually the case. Since Eq. 33 does not include inertia, the condition for the inertia neglecting would be received by compare to systems of SA/LP and gas.

7.1 Bending-Dominated Rate

First order approximation

When Eq. 33 is expanded in small Ca, the term $\alpha_1 + \alpha_2$ is found to be bound from above by $|q|\sqrt{\Theta}$. To first order in small Ca and large Θ , a solution can be obtained by taking De_2^{-1} to one. This solution is:

$$\omega = \omega_K = -\frac{iKq^2}{\eta_M} \quad (53)$$

This is again the pure undulation bulk mode as described in subsection 6.1 and in appendix B.

Bending-Dominated Rate with Correction due to Surface tension

Trying to get corrections for the bending bulk mode should be done by adding components from outside the brackets in Eq. 33: In the limit of $Ca \ll 1$ there is no solution to second order approximation. In the limit of $Ca \gg 1$ a solution is achieved.

The approximation considered are:

$$Ca \gg s, \quad De_1^{-1} \ll \min\{Ca\sqrt{\Theta}, \Theta\}, \quad De_2^{-1} \ll \Theta, \quad Re \ll 1 \quad (54)$$

In the condition for Ca we use $\alpha_1 \simeq |q|\sqrt{\Theta} \gg \alpha_2$; we shall use this also in the approximation for Eq. 33:

$$Ca\sqrt{\Theta} - \sqrt{1 - De_2^{-1}\Theta} = 0$$

This equation is the same as Eq. 41 derived for SA/LP and gas. Its solution is $\omega = \omega_{K,(\gamma)}$ (Eq. 42), which gives the surface mode discussed in subsection 6.1. The difference between 6.1 and this case is the extra approximation needed here: $Ca \gg s$. This approximation affects the previous relevant $|q|$ range, Eq. 44, by adding the restriction: $|q| \ll \frac{\gamma\eta_M}{K\eta}$, which is practically always fulfilled.

7.2 Compression-Dominated Rate

First Order Approximation

Expanding Eq. 33 in small capillary number assuming $De_2^{-1} \ll De_1^{-1}$ leads to the solution:

$$\begin{aligned} \omega = \omega_B &= -\frac{iB}{2(\eta_T + \eta_V)} \\ \alpha_{1,2} &= |q|\sqrt{-1 \pm \sqrt{De_2^{-1}}} \end{aligned} \quad (55)$$

This is a bulk, not a surface mode, see subsection 6.2 and appendix C.

Compression and Bending Dominated Rate

For $De_2^{-1} \gg 1$, and the additional assumptions of $Ca \ll \Theta De_2^{-1}$, Eq. 33 takes the form:

$$-De_1^{-1} - De_2^{-1} + \Theta = 0$$

The solution for this is:

$$\omega = \omega_B = -\frac{i(B + Kq^2)}{2(\eta_T + \eta_V)} \quad (56)$$

The relevant $|q|$ range in which this is a surface mode are essentially the same as the one deduced in the case of SA/LP and gas, described in 6.2.

7.3 Surface Tension Dominated Rate

First Order Approximation

Under the approximations:

$$\text{De}_1^{-1} \ll \Theta, \quad \text{De}_2^{-1} \ll 1, \quad \text{Ca} \gg 1$$

The determinant equation (Eq. 33) is approximately:

$$-\text{Ca}\sqrt{\Theta} + \Theta = 0$$

This gives the surface mode of ω_γ presented in 6.3.

The correction for the surface tension dominated rate due to bending and compression can be achieved easily, in essentially the same way as for systems of SA/LP and gas.

Simple Fluid Correction for the Surface Tension Dominated Mode

More terms that include the surface tension appear when we examine Eq. 33 under the approximations:

$$\text{De}_1^{-1} \ll \sqrt{\Theta}, \quad \text{De}_2^{-1} \ll \sqrt{\Theta}^{-1} \quad (57)$$

The determinant equation (Eq. 33) is then reduced to:

$$-\text{Cas} - \text{Ca}\sqrt{\Theta} + 2s\sqrt{\Theta} + \Theta = 0$$

The temporal decay of the surface mode, deduced using the perturbation theory, is:

$$\omega = \omega_{\gamma(s)} = -\frac{i\gamma|q|}{\sqrt{2\eta_M(\eta_T + \eta_V)}} - \frac{i\gamma|q|\eta}{\eta_M(\eta_T + \eta_V)}$$

The relevant $|q|$ range for this solution should be, according to Eq. 57 and the condition $\text{Re} \ll 1$, is given by (compare to subsection 6.3):

$$\max\left\{\frac{\gamma\rho}{\eta_M^2\sqrt{\Theta}}, \frac{B}{\gamma}\right\} \ll q \ll \frac{\gamma}{\Theta K}$$

The conditions on the system turn out not to be constraining physically, as in subsection 6.3.

7.4 Large Simple Fluid Viscosity Limit

The parameter s is proportional to the simple fluid viscosity. For large viscosity of the simple fluid $s \rightarrow \infty$. Expanding Eq. 33 in large s gives the equation:

$$s^2 - \text{Cas} + \frac{\alpha_1 + \alpha_2}{|q|}s + \sqrt{1 - \text{De}_2^{-1}}\frac{\alpha_1 + \alpha_2}{|q|}s = 0$$

where De_1^{-1} enters in this case only through α_1, α_2 . For $\text{De}_1^{-1} \ll \Theta$, $\text{De}_2^{-1} \ll 1$ this equation is approximately $s^2 - \text{Cas} + 2s\sqrt{\Theta} = 0$. By returning to dimensionless parameters, this equation reaches the form:

$$\frac{i\gamma|q|}{\omega} + 2\eta + 2\sqrt{2\eta_M(\eta_T + \eta_V)} = 0$$

The solution for this approximation is therefore

$$\omega = -\frac{i\gamma|q|}{2\eta + 2\sqrt{2\eta_M(\eta_T + \eta_V)}}$$

This solution is, similar to the one for two simple fluids Eq. 25, where the SA/LP viscosity enters as $\sqrt{2\eta_M(\eta_T + \eta_V)}$, just as in ω_γ (6.3).

8 Inclusion of Slip

In this chapter we include the slip property, that is specific for lamellar phase (and does not exist in single-component smectic A).

The Determinant Equations

The lamellar bulk equations (Eq. 11- 16), including the slip property (in addition to the properties common to all smectics A) yield the following relations between the hydrodynamic variables:

$$\begin{aligned} v_x &= \frac{i}{q} \frac{\partial v_z}{\partial z} \\ u &= \frac{i}{\omega} v_z \\ p &= (\eta_M - \frac{1}{\mu q^2} - 2\eta_T) \frac{\partial v_z}{\partial z} + \frac{\eta_M}{q^2} \frac{\partial^3 v_z}{\partial z^3} \\ v_{L,x} &= 0 \\ \psi &= -\frac{1}{q^2 \mu} \frac{\partial v_z}{\partial z} \end{aligned}$$

The resulting equation for v_z gives the spatial decays:

$$\frac{\alpha_{1,2}}{|q|} = \sqrt{\frac{1}{2}\Theta + \frac{1}{2}m - 1 - \frac{1}{2}\text{De}_1^{-1} - \frac{1}{2}\text{Re} \pm \sqrt{\Upsilon}}$$

where

$$\Upsilon = \left(\frac{1}{2}\Theta + \frac{1}{2}m - 1 - \frac{1}{2}\text{De}_1^{-1} - \frac{1}{2}\text{Re}\right)^2 - (1 - \text{De}_2^{-1} - \text{Re})$$

m is a positive dimensionless parameter containing the slip coefficient, $m = (\mu\eta_M q^2)^{-1}$.

We see that the change in the spatial decays, compared to SA/LP spatial decays (Eq. 29), is obtain by taking $\Theta \rightarrow \Theta + m$. Following the boundary conditions, for the case of lamellar phase and gas, the same modification occurs also in the determinant equation. The determinant equation for system of lamellar phase and gas changes to:

$$\text{Ca} \frac{\alpha_1 + \alpha_2}{|q|} + \text{Re} - \sqrt{1 - \text{De}_2^{-1} - \text{Re}(\Theta + m - \text{De}_1^{-1} - \text{De}_2^{-1} - \text{Re})} = 0 \quad (58)$$

In the case of lamellar phase and simple fluid, the boundary condition for the tangent velocity is changed. When slip is included, there are two tangent velocities characterizing

the lamellar phase: the average tangent velocity v_x , and the surfactant fraction tangent velocity $v_{L,x}$. Taking the extreme bilayer as the interface, we demand that the simple fluid's tangent velocity is equal to the surfactant fraction's tangent velocity.

Following this condition, the determinant equation for the lamellar phase and simple fluid changes significantly from Eq. 33. The current equation is:

$$(\text{Ca} - s) \frac{\alpha_1 + \alpha_2}{|q|} - \sqrt{1 - \text{De}_2^{-1}} (\Theta + m - \text{De}_1^{-1} - \text{De}_2^{-1}) = 0 \quad (59)$$

(Inertia has been omitted here for the reasons detailed in 5.3).

9 Slip Influenced Decay Rates

For $\text{Ca} \gg s$ the dynamics for lamellar phase and gas is similar to that of lamellar phase and simple fluid. In this section we derive the surface modes for lamellar phase and gas according to Eq. 58. Placing the solutions for ω in Ca shows that all these modes obey the condition $\text{Ca} \gg s$ (for $s \simeq 1$). Therefore, they are relevant as well to systems of lamellar phase and simple fluid, when the simple fluid's viscosity is similar to that of the solvent (water).

Since the change in the rate equation can be done by replacing Θ with $\Theta + m$, the surface modes can be obtained easily by changing the surface modes found for SA/LP in the same way. We study in this section the influence of the slip property by considering the cases when it is more dominant than the large viscosities represented by Θ , i.e. we shall assume $m \gg \Theta$. Since $m \sim q^{-2}$ and $\Theta \sim q^0$, this assumption is always valid for sufficiently small wave numbers, $|q| \ll (\eta_M \mu)^{-1/2} \Theta^{-1/2}$. As discussed in subsection 2.4, $(\eta_M \mu)^{1/2}$ is approximately the distance between layers. Therefore this condition is easily fulfilled. As in the previous sections, we are interested in processes that are not affected by inertia.

9.1 Bending-Dominated Rate

First Order Approximation

The pure undulation bulk mode $\omega = \omega_K = -\frac{iKq^2}{\eta_M}$ is achieved, as in subsection 6.1, by equating De_2^{-1} with one. Therefore the rate of this mode is not affected by adding the slip property.

Correction due to Surface Tension

In order to achieve a correction for the bending bulk mode due to surface tension, we look at the approximate version of Eq. 58 while including the surface tension element:

$$\text{Ca}\sqrt{m} - \sqrt{1 - \text{De}_2^{-1}m} = 0$$

The surface mode corresponding to this has the spatial decays:

$$\alpha_1 = (\mu\eta_M)^{-1/2}, \quad \alpha_2 = \frac{\gamma(\mu\eta_M)^{3/4}}{K}q^{3/2}$$

And the temporal decay:

$$\omega = \omega_{K,(\gamma)}^s = -\frac{iKq^2}{\eta_M} - \frac{i\gamma^2\mu q^2}{K}$$

The ratio between the spatial decays' amplitudes is given by Eq. 30. In this case it is approximately $C_1\alpha_1^2 = -C_2\alpha_2^2$. Substituting the above spatial decays we can see that $|C_2| \gg |C_1|$. This correction is based on the assumptions:

$$m \gg \Theta, \quad m \gg Ca\sqrt{m} \gg \max\{\text{Re}, \text{De}_1^{-1}, \Theta\}, \quad \text{De}_1^{-1} \ll m, \quad \text{Re} \ll 1$$

The conditions on $|q|$ are therefore:

$$|q| \ll \min\{(\sqrt{2\mu(\eta_T + \eta_V)})^{-1}, \frac{1}{K}(\frac{\gamma^2\eta_M^3}{\mu\rho^2})^{1/4}, \frac{\gamma}{\Theta K\sqrt{\mu\eta_M}}\}$$

And the resulting conditions on system parameters are:

$$\frac{B\mu\eta_M}{K} \ll 1, \quad \frac{B^2\mu\eta_M}{\gamma^2} \ll 1, \quad \frac{\gamma\sqrt{\eta_M\mu}}{K} \ll 1$$

This is a diffusive surface mode ($\omega \sim q^2$). From the conditions on the system this mode is valid for systems with high bending modulus and small compression modulus. From the conditions on $|q|$ we can see that this mode is valid for a wide range of wave numbers, restricted only from above. The spatial decay length given by α_1 is typically of the order of the microscopic distance between layers. Therefore, this spatial decay exists only within a thin boundary layer. However, the dominant spatial decay α_2 gives a typical decay length larger than the wave number.

9.2 Compression-Dominated Rate

First Order Approximation

The compression bulk mode with ω_B was achieved in subsection 6.2, by equating De_1^{-1} with Θ . This bulk mode is affected by the slip property since the equality, in the case $m \gg \Theta$, is now between De_1^{-1} and m :

$$\omega = \omega_B^s = -iB\mu q^2$$

Correction due to Bending

Expanding the above result for first order in De_2^{-1} , based on the approximate version of Eq. 58:

$$m - \text{De}_1^{-1} - \text{De}_2^{-1} = 0$$

gives, as in 6.2, one spatial decay ($C_2 = 0$):

$$\alpha = |q| \sqrt{\text{De}_2^{-1} - 1} = |q| \sqrt{\frac{K}{\eta_M \mu (B + Kq^2)} - 1}$$

and the temporal decay:

$$\omega = \omega_{B,K}^s = -i(Bq^2 + Kq^4)\mu$$

This correction is based on the assumptions:

$$\text{Ca} \frac{\alpha}{|q|} \ll \text{De}_2^{-1} \sqrt{\text{De}_2^{-1} - 1}, \quad \text{De}_2^{-1} \gg \Theta, \quad \text{Re} \ll \text{De}_2^{-1} \sqrt{\text{De}_2^{-1} - 1}, \quad m \gg \Theta$$

The conditions on $|q|$ are therefore:

$$\frac{\gamma}{K} \ll |q| \ll (\sqrt{2\mu(\eta_T + \eta_V)})^{-1}$$

The lower limit restricts q to very high values, at the edge of validity of the hydrodynamic theory. The resulting conditions on system parameters are:

$$\frac{K}{2\mu B(\eta_T + \eta_V)} \gg 1, \quad \frac{\rho B K}{\eta_M} \ll 1, \quad \frac{\gamma \sqrt{2\mu(\eta_T + \eta_V)}}{K} \ll 1$$

The most problematic condition here is the restriction on $\min |q|$, restricted to neglect of the surface tension. For $\gamma \simeq k_B T/d^2$, $K \simeq k_B T/d^3$ it yields $|q| \gg d^{-1}$. This surface mode is therefore valid only in systems with extremely low surface tension, high bending modulus and low compressibility.

9.3 Surface Tension Dominated Rate

First Order Approximation

The surface tension dominated rate was achieved in subsection 6.3 by equating Ca with $\sqrt{\Theta}$. This surface mode is therefore affected by the slip property. In the case $m \gg \Theta$, Ca should

be equated with \sqrt{m} . The temporal decay changes to:

$$\omega = \omega_\gamma^s = -i\gamma\sqrt{\frac{\mu}{\eta_M}} q^2$$

The spatial decays are then:

$$\alpha_1 = (\sqrt{\mu\eta_M})^{-1}, \quad \alpha_2 = \sqrt{\mu\eta_M}q^2$$

Since $m \gg \Theta \gg 1$ the spatial decays obey $\alpha_2 \gg \alpha_1$. From Eq. 30 the amplitude ratio is achieved, we get $|C_2| \gg |C_1|$.

This case is based on the assumptions:

$$m \gg \text{De}_1^{-1}, \quad \text{De}_2^{-1} \ll 1, \quad \text{Re} \ll 1, \quad m \gg \Theta$$

The condition on $|q|$ is therefore:

$$|q| \ll (\sqrt{2\mu(\eta_T + \eta_V)})^{-1}$$

and the conditions on the system are:

$$\frac{B\sqrt{\eta_M\mu}}{\gamma} \ll 1, \quad \frac{\gamma\sqrt{\eta_M\mu}}{K} \gg 1, \quad \frac{\gamma\rho\sqrt{\mu}}{\sqrt{\eta_M^3}} \ll 1$$

All the above conditions are possible for lamellar phase typical parameters. The exception is the condition $B\sqrt{\eta_M\mu}/\gamma \ll 1$, which is only marginally fulfilled. This is therefore a diffusive surface mode ($\omega \sim q^2$), valid mainly in concentrated lamellar phases (since $\mu \sim d^2$), with large enough surface tension. The dominant spatial decay is α_2 , which gives a decay length much larger than the wave length, $\alpha_2^{-1} \sim (dq^2)^{-1}$. The spatial decay length given by α_1 is of the order of the distance between layers; it therefore describes a thin boundary layer.

In contrast with the surface tension dominated mode where the dissipative mechanism is viscosity, sec. 6.3, the validity of this surface mode is not limited by inertia, (this is because both m and Re are $\propto q^2$). The significant restriction is due to the omission of elastic forces. Moreover, also for the elasticity-dominated surface modes, subsections 9.1 and 9.2, inertia does not create a significant restriction. The limiting condition in all these asymptotic cases derive from the restoring forces not included in each case. In subsection 9.5 we eliminate these restrictions by considering all restoring forces.

Correction due to Bending

An approximate version of Eq. 58, for first order in small De_2^{-1}

$$\text{Ca}\sqrt{m} - \sqrt{1 - \text{De}_2^{-1}m} = 0$$

gives the following correction to the above surface mode. The temporal decay for this correction is:

$$\omega = \omega_{\gamma(K)}^s = -i\gamma\sqrt{\frac{\mu}{\eta_M}}q^2 - \frac{iKq^2}{2\eta_M}$$

Correction due to Compression

The approximate version :

$$\text{Ca}\sqrt{m - \text{De}_1^{-1}} - (m - \text{De}_1^{-1}) = 0$$

of Eq. 58 gives a surface mode with the temporal decay:

$$\omega = \omega_{\gamma(B)}^s = -i\left(\gamma\sqrt{\frac{\mu}{\eta_M}} + \frac{1}{2}B\mu\right)q^2$$

Correction due to Lamellar Viscosity

In order to consider the effect of the lamellar viscosity in addition to the slip effect in the surface tension dominated rate, we solve the following approximation for Eq. 58:

$$\text{Ca}\sqrt{m + \Theta} - (m + \Theta) = 0$$

This gives the temporal decay:

$$\omega = \omega_{\gamma(\Theta)}^s = -i\gamma\sqrt{\frac{\mu}{\eta_M}}q^2 + i\gamma\mu^{3/2}\eta_M^{-1/2}(\eta_T + \eta_V)q^4$$

Correction due to Simple Fluid Viscosity

For simple fluid with large viscosity, a correction to ω_γ^s can be obtained. By considering s in the approximation to Eq. 59 we have:

$$(\text{Ca} - s)\sqrt{m} - m = 0$$

The correction achieved is:

$$\omega = \omega_{\gamma(\eta)}^s = -i\gamma\sqrt{\frac{\mu}{\eta_M}}q^2 + 2i\gamma\frac{\mu}{\eta_M}\eta q^3$$

The conditions on q and on the system are essentially the same, for all the corrections for the surface tension dominated rate, as in the first order approximation.

9.4 Large Simple Fluid Viscosity

If the simple fluid viscosity is extremely large, the surface relaxation is governed by the simple fluid phase. In this case Eq. 59 is approximately:

$$(\text{Ca} - s)\sqrt{m} = 0$$

Therefore

$$\omega = -\frac{i\gamma|q|}{2\eta}$$

as on the surface of a simple fluid.

9.5 Summary

We have seen that including the slip property reveals a different relaxation, all of whose asymptotic rates obey a diffusive law, $\omega \sim q^2$. The bending-dominated surface mode was achieved by a correction due to surface tension. It exhibits a boundary layer (given by α_1), and has one dominant decay length (given by α_2). This mode is allowed in systems where the compressibility is sufficiently small.

The compression and bending dominated mode is relevant for very small surface tension, high bending modulus and low compressibility. The surface tension neglect restricts the allowed wave numbers from below to a small permitted region. This mode has one spatial decay.

The asymptotic approximation for large surface tension, section 9.3, gives the surface tension dominated mode. In this mode there are two spatial decays. The first yields a boundary layer. The second, dominant spatial decay is $\alpha_2 = \sqrt{\eta_M\mu}q^2$. This gives a typical

decay length much larger than the wave length, (i.e., larger than the simple fluid decay length). This mode is valid for reasonably low elastic moduli and high surface tension coefficient. We summarize these results in Table 2.

The main validity restriction on each of these solutions is due to neglecting the other possible restoring forces. (For example, the surface tension dominated mode is restricted by the lamellar elastic forces. Nevertheless, it is not restricted because of inertia.) Therefore, the real surface relaxation will involve all these mechanisms. As we saw above, when the dominant dissipative mechanism is slip ($m \gg \Theta$), the decay rates are quadratic in the wave number $\omega \sim q^2$. To achieve more realistic expressions for the slip-influenced surface mode we thus study Eq. 59 assuming such a quadratic dependence. We shall also neglect 1 and Θ with respect to m . We also consider the case where inertia is negligible, $\text{Re} \ll 1$. By that, the spatial decays satisfy: $\frac{\alpha_1 + \alpha_2}{|q|} \simeq \frac{\alpha_1}{|q|} \simeq \sqrt{m - \text{De}_1^{-1}}$, and the determinant equation has the form:

$$\text{Ca} \sqrt{m - \text{De}_1^{-1}} - \sqrt{1 - \text{De}_2^{-1}}(m - \text{De}_1^{-1}) = 0$$

Substituting $\omega = -iAq^2$, we find A to be:

$$A = \frac{1}{2\eta_M} (B\mu\eta_M + K \pm \sqrt{(B\mu\eta_M - K)^2 + 4\eta_M\mu\gamma^2}) \quad (60)$$

which gives two modes:

$$\omega = \frac{1}{2} \left(\omega_b^s - \omega_K \pm \sqrt{(\omega_b^s + \omega_K)^2 + (2\omega_\gamma^s)^2} \right)$$

The \pm should be taken so $\text{De}_1^{-1} < m$ and $\text{De}_2^{-1} < 1$, which gives $A > B\mu$ and $A > K/\eta_M$ respectively. Note that this derivation gives bulk modes for $\gamma = 0$, as seen in subsections 9.1 and 9.2. It is also important to see that we assumed $\alpha_1 \gg \alpha_2$ and therefore this solution becomes invalid for $\text{De}_1^{-1} \rightarrow m$. (The case $\text{De}_1^{-1} \rightarrow m$ was referred to in subsection 9.2.)

Since $\alpha_1 \gg \alpha_2$ we get from the boundary conditions Eq. 17 that $C_1 \ll C_2$; the dominant spatial decay is therefore

$$\alpha_2 = |q| \sqrt{\frac{1 - \text{De}_2^{-1}}{m - \text{De}_1^{-1}}} = q^2 \sqrt{\frac{\mu(\eta_M A - K)}{A - B\mu}} \quad (61)$$

	surface tension dominated relaxation
decay rate	$\omega = \omega_\gamma^s = -i\gamma\sqrt{\frac{\mu}{\eta_M}} q^2$
spatial decays	$\alpha_1 = (\sqrt{\mu\eta_M})^{-1}, \quad \alpha_2 = \sqrt{\mu\eta_M}q^2$
$ q $ range	$ q \ll (2\mu(\eta_T + \eta_V))^{-1/2}$
system conditions	$\frac{B\sqrt{\eta_M\mu}}{\gamma} \ll 1, \quad \frac{\gamma\sqrt{\eta_M\mu}}{K} \gg 1, \quad \frac{\gamma\rho\sqrt{\mu}}{\sqrt{\eta_M^3}} \ll 1$
remarks	valid for low compression modulus
	compression-dominated relaxation
decay rate	$\omega = \omega_{B,K}^s = -i(Bq^2 + Kq^4)\mu$
spatial decay	$\alpha = q \sqrt{\frac{K}{\eta_M\mu(B+Kq^2)}} - 1$
$ q $ range	$\frac{\gamma}{K} \ll q \ll (2\mu(\eta_T + \eta_V))^{-1/2}$
system conditions	$\frac{K}{2\mu B(\eta_T + \eta_V)} \gg 1, \quad \frac{\rho BK}{\eta_M} \ll 1, \quad \frac{\gamma\sqrt{2\mu(\eta_T + \eta_V)}}{K} \ll 1$
remarks	valid only for extremely low surface tension
	bending-dominated relaxation
decay rate	$\omega = \omega_{K,(\gamma)}^s = -\frac{iKq^2}{\eta_M} - \frac{i\gamma^2\mu q^2}{K}$
spatial decays	$\alpha_1 = (\sqrt{\mu\eta_M})^{-1}, \quad \alpha_2 = \frac{\gamma(\mu\eta_M)^{3/4}}{K}q^{3/2}$
$ q $ range	$ q \ll (2\mu(\eta_T + \eta_V))^{-1/2}$
system conditions	$\frac{B\mu\eta_M}{K} \ll 1, \quad \frac{B^2\mu\eta_M}{\gamma^2} \ll 1, \quad \frac{\gamma\sqrt{\eta_M\mu}}{K} \ll 1$
remarks	valid for low compression modulus and high bending modulus

Table 2: Results - surface modes influenced by slip

Equations 60 and 61 are the central result of this work. They describe a diffusive surface mode affected by all restoring forces and damped by the slip mechanism. It governs the surface relaxation over a wide range of wave lengths.

10 Discussion

In this study we aimed to define the surface modes characterizing the relaxation of an interface between a lamellar phase and gas or simple fluid. We have studied the modes emerging from the features of simple (thermotropic) smectic A, and the modes emerging from addition of the slip property that is specific to the lyotropic lamellar phase.

For simple smectic A liquid crystal we have shown that there are two asymptotic solutions, one governed by surface tension and the other by the elastic features of the material. The mode governed by elasticity (B, K) is unlikely to be manifested in practice, being restricted to too small wave lengths.

The surface tension dominated relaxation is more likely to be manifested. This mode however is also limited to small wave lengths because of inertia. It resembles the surface mode of a simple fluid. The decay rate converges to that of a simple fluid if we take the smectic viscosity coefficients to be equal. This mode has two very different spatial decays due to the anisotropic viscosity. The dominant decay length is an order of magnitude larger than the one for a simple fluid.

Thus, if the inertia is negligible the surface relaxation should be dominated by surface tension, as has been predicted also by the Orsay group [2]. The Orsay group has predicted, as mentioned in the introduction, a surface mode similar to that of simple fluid, but with a modified surface tension due to a known static contribution from elasticity [4], $\gamma \rightarrow \gamma + \sqrt{BK}$. Our results do not show such a correction in the dynamic description of surface relaxation.

Since the above modes for thermotropic smectic A are relevant only for very small wave lengths, the mechanism underlying the relaxation for more realistic q should involve inertia. This conclusion of ours agrees with the analysis of Fedorov et al [1].

The major new contribution of the current work is the inclusion of the slip mechanism in the analysis of surface relaxation. We have discovered that when the dissipation in the system is governed by the slip mechanism, which is an assumption valid over a wide range of wave lengths, there are slowly decaying surface modes that do not involve (or are restricted by) inertia. The slip property of a lamellar phase stems from its two-component structure.

This property does not exist in thermotropic smectic A.

The bending-dominated surface relaxation involves also surface tension, $\omega = \omega_{K,(\gamma)}^s = -\frac{iKq^2}{\eta_M} - \frac{i\gamma^2\mu q^2}{K}$. It has a boundary layer whose width is of the order of the distance between layers. The typical decay length is given by $\alpha_2 = \frac{\gamma(\mu\eta_M)^{3/4}}{K}q^{3/2}$. This mode is relevant for systems with low compressibility and a high bending modulus. It can appear over a large range of wave lengths.

The mode dominated by the compressibility of the lamellar phase has $\omega = \omega_{B,K}^s = -i(Bq^2 + Kq^4)\mu$. It is limited to very small wave lengths. It is relevant for systems with negligible surface tension, high bending modulus and low enough compression modulus.

The relaxation governed by surface tension is predominant. As in the bending and compressibility dominated cases, it is a diffusive mode, $\omega \sim q^2$. In contrast to the mode governed by surface tension for smectic A, this mode is very different from the one manifested for a simple fluid. The difference results from the change in the dominant dissipative mechanism. The decay rate is given by $\omega = -i\gamma\sqrt{\frac{\mu}{\eta_M}}q^2$. This mode has two spatial decay coefficients: $\alpha_1 = (\sqrt{\eta_m\mu})^{-1}$ and $\alpha_2 = \sqrt{\eta_m\mu}q^2$. α_1 gives a short decay length of the order of the distance between layers. This is therefore only a boundary layer. The dominant spatial decay is given by α_2 , yielding a decay length much larger than the wave length (the wave length gives the decay length for a simple fluid). This mode is valid for wave lengths larger than $\sim 10d$, which is also the validity range of the entire hydrodynamic description.

We have been able to interpolate the various asymptotic cases by assuming a diffusive rate, $\omega \sim q^2$, whose coefficient depends on all three possible restoring mechanisms (surface tension, bending and compression). This central result is summarized in Equations 60,61, yielding two diffusive modes. Our analysis shows that these two slip-damped diffusive modes, unique to lyotropic lamellar phase, should govern the surface relaxation over a wide range of wave lengths. For the simplest case of sterically stabilized lamellar phases, characterized by a single energy scale, $k_B T$, and a single length scale, d , these modes have a decay rate of $\sim \frac{k_B T}{\eta_M d}q^2$ and a very large penetration depth, $\sim (dq^2)^{-1}$.

To our best knowledge, the surface relaxation of lyotropic lamellar phases has not yet been studied experimentally. Our predictions, which qualitatively deviate from those for

other liquids, could be checked using scattering techniques.

The present findings encourage further research aimed to elucidate the intriguing, poorly understood, phenomenon of myelin figures forming at non equilibrium interfaces between lamellar phases and simple fluids. At the interface, finger like structures emerge, composed of the bilayers of the lamellar phase. We briefly describe the myelin figure phenomenon and suggest preliminary hydrodynamic consideration for it in appendix D.

APPENDICES

A Rejecting the Possibility of Only One Spatial Decay in Bulk SA/LP Hydrodynamics

In this appendix we examine the case where the velocity $v_z(z)$, given by Eq. 28 of the lamellar bulk hydrodynamics, has only one spatial decay length, as in a simple fluid, and therefore $v_z(z)$ has the form $v_z(z) = (a+bz) \exp(\alpha z)$. Substituting this solution in Eq. 28, we compare terms having the same power of z . The equation given by terms of z^1 is the same one obtained from assuming two spatial decays. In order to fulfill the assumption of only one spatial decay the inner root in the solution for α , Eq. 29, should vanish, so that

$$\alpha = |q| \sqrt{\frac{1}{2}\Theta - 1 - \frac{1}{2}\text{De}_1^{-1} - \frac{1}{2}\text{Re}}$$

This demand sets $\omega(q)$. However, the resulting $\omega(q)$ does not satisfy the boundary equations. Therefore, it cannot describe a surface mode.

B Pure Undulation Bulk Mode

Taking $q_z = 0$ and $q_x \equiv q$, the hydrodynamic variables of the SA/LP have a spatial dependence of $\exp(iqx)$. Neglecting inertia, the bulk equations for SA/LP, Eq. 28, give the temporal decay:

$$\omega = -\frac{iKq^2}{\eta_M}$$

This is called the undulation bulk mode [8, 3].

In this mode the distance between layers is not modulated ($q_z = 0$), the pressure turns out to be zero, and the oscillating velocity is only in the z direction.

C Compression Bulk Mode

Taking $q_x = q_z \equiv q$ the hydrodynamic variables of the SA/LP have a spatial dependence of $\exp(iqx)\exp(iqz)$. Neglecting inertia, the bulk equations for SA/LP, Eq. 28, give the temporal decay:

$$\omega = -\frac{i(B + Kq^2)}{2(\eta_V + \eta_T)} \simeq -\frac{iB}{2(\eta_V + \eta_T)}$$

The typical decay time for this bulk mode is quite fast, $|\omega|^{-1} \sim \eta_T/B \sim 10^{-4} - 10^{-8}\text{sec}$. The neglect of inertia is valid for:

$$|q| \gg \frac{\sqrt{\rho B}}{\eta_T + \eta_V} \sim 10^3\text{m}^{-1}$$

This condition is obtained by comparing the inertial term with the term that involves compression.

D Myelin - Suggestion for Future Research

D.1 Description of the Myelin Phenomenon

When a lamellar phase comes in contact with its solvent, and the two phases are out of equilibrium, the solvent will penetrate into the lamellar phase. During the penetration of the solvent into the lamellar phase, as it becomes progressively swollen, “myelin” figures may appear on the interface between the simple fluid (the solvent) and the lamellar phase. Myelin figures look like fingers reaching out from the lamellar phase toward the solvent. The width of the myelin figures is about $10\ \mu\text{m}$ and their length is up to $150\ \mu\text{m}$. Myelin figures are named after the myelin that wrap the extensions of nerve cells - the axons. The neural myelin has an extremely important role in the conduction of electric signals throughout the axons. Thus myelin contains multi-layers of lipid both in nerve axons and in lamellar phases. The phenomenon of myelin figure formation was discovered in 1854 by Virchow [15].

Quantitative descriptions of myelin figures’ formation appear in recent experiments by Buchanan et al [16, 17]. These works found that myelin figures appear about 20 seconds after the creation of contact between the lamellar phase and its solvent. It was found that

for myelin formation to occur it is necessary that the surfactant does not dissolve easily in the solvent. According to Buchanan and colleagues [16], during the process of myelin figures formation, the length of the myelin fingers is proportional approximately to the square root of the time elapsed from the moment of contact with the solvent. This implies the involvement of diffusion.

Each “myelin finger” is a cylindrical structure, made of many bilayers. Approximate calculation of the number of bilayers in each “myelin finger” can be reached by dividing the width of a “finger” ($10 \mu\text{m}$) by the characteristic distance between layers (100 \AA). This yields thousands of layers in each “finger”.

D.2 Basic Hypotheses for Modelling of Myelin Figures’ Formation

Imagine a lamellar phase in the area $z < 0$, where the normal to its layers is oriented in the z direction, and its solvent is in the area of $z > 0$. A pressure gradient makes the solvent penetrate into the lamellar phase. As a consequence, the lamellar phase swells in the interface area. In this state the system is out of equilibrium. Modelling the interface distortion occurring upon the penetration of the solvent into the lamellar phase, a description based on a set of sinusoidal distortions (Fourier Transform) can be used. Considering the linear properties of FT an arbitrary sinusoidal wave length can be treated, as we have done in the surface mode analysis.

When such a distortion is formed, will it fade (with the system regaining a flat interface) or will the amplitude of the distortion increase due to persistent instability? Assuming sinusoidal distortion in which the interface shape is unstable, the amplitude of the distortion increases progressively. After sufficient time the interface shape will look like fingers, the width of each is related to the unstable wave length. One objection to this description is that it is not known whether the lamellar layers are in all cases parallel to the interface.

Thus, we ask whether a hydrodynamic model can be constructed that allows for an unstable interface in which the distortion increases progressively without fading. If so, we ask whether a correlation can be shown between the width of the myelin fingers formed and the wave length of the unstable sinusoidal distortion.

The difference between this model and the model used for surface modes is that surface modes are perturbations on equilibrium state, whereas the current model refers to perturbations on a steady flow (where the system is out of equilibrium). The steady flow is uniform in the x, y coordinates and does not change in time.

The steady flow properties depend on the position along the z direction. Essentially, the values of all hydrodynamic coefficients $B, K, \mu..$ depend on the distance between layers d , which is changed due to the steady penetration flow. Therefore, the steady flow can make the hydrodynamic equations non-linear.

The bulk dynamics is define by the bulk equations of the lamellar and the simple fluid phases Eqs. 11-16 and Eqs. 6-7, and the boundary conditions Eqs. 17-21. However, we should exchange d in the expressions of these parameters to $d_0(1 + \frac{\partial u}{\partial z})$. For example, if $B \sim d^{-3}$, we have $B \frac{\partial^2 u}{\partial z^2} \simeq B_0(1 - 3\frac{\partial u}{\partial z}) \frac{\partial^2 u}{\partial z^2} \simeq B_0(1 - 3\frac{\partial u_h}{\partial z}) \frac{\partial^2 \tilde{u}}{\partial z^2} + B_0(1 - 3\frac{\partial \tilde{u}}{\partial z}) \frac{\partial^2 u_h}{\partial z^2}$, where \tilde{u} is the perturbation and u_h is given by the steady flow. Thus, the dynamics are described by non-linear equations, which are linearized in the small perturbation. Such additional term in the hydrodynamic equation may cause the interface to be unstable.

In the next subsection we derive the steady flow and discuss the sugested model.

D.3 Solution for a Steady Flow

The steady flow represents the solution for a stable system that is uniform in the directions parallel to the layers. In this case the bulk hydrodynamic equations (Eq. 11-16) reduce to the form:

$$\begin{aligned} 0 &= \frac{\partial v_z}{\partial z} \\ 0 &= -\frac{\partial p}{\partial z} + B \frac{\partial^2 u}{\partial z^2} + 2\eta_V \frac{\partial^2 v_z}{\partial z^2} \\ v_z &= -\lambda_p B \frac{\partial^2 u}{\partial z^2} \end{aligned} \tag{62}$$

The permeation process must be included here in order to achieve a non trivial solution for the steady flow (in contrast with the surface modes derivation).

The simple fluid bulk hydrodynamics (Eq. 6-7) is given by:

$$\begin{aligned} 0 &= \frac{\partial v_z}{\partial z} \\ 0 &= -\frac{\partial p}{\partial z} + 2\eta \frac{\partial^2 v_z}{\partial z^2} \end{aligned} \tag{63}$$

The interfacial boundary conditions that are relevant in this case are the continuity of the velocity v_z , the discontinuity in the force exerted on the interface because of the surface tension, and the pressure continuity that is possible due to permeation. Apart from that we demand that the lamellar layers be fixed at a distance L from the interface: $u(z = -L) = 0$, and set a pressure difference Δp between $z = L$ in the simple fluid side and $z = -L$ in the lamellar side. Since the interface remains flat in a steady flow the surface tension's contribution is zero.

The hydrodynamics of the steady flow is then:

$$\begin{aligned} v_{z,\text{SF}} &= v_{z,\text{LP}} = -\lambda_p \frac{\Delta p}{L} \\ p_{\text{LP}} &= \frac{\Delta p}{L} z + p_L \\ p_{\text{SF}} &= p_L \\ u &= \frac{\Delta p}{2LB} (z^2 - L^2) \end{aligned} \tag{64}$$

The subscripts ‘‘SF’’ and ‘‘LP’’ represent ‘‘Simple Fluid’’ and ‘‘Lamellar Phase’’ respectively.

In order to receive the linear instability, the steady flow plus the perturbation should be substituted in the non-linear equations. These equations should then be linearized in the perturbation. The solution for ω shows growth if $i\omega < 0$, and decay if $i\omega > 0$. The instability, meaning decay that transfers into growth, should become more dominant as the driving force Δp increases.

Presenting the open question about the creation of myelin figures, we encourage a further research that could be based on the equilibrium study of surface modes in lamellar phase presented in this work, and the out of equilibrium model suggested in this appendix.

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