Layered Systems Under Shear Flow

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Abstract We discuss and review a generalization of the usual hydrodynamic description of smectic A liquid crystals motivated by the experimentally observed shear-induced destabilization and reorientation of smectic A like systems. We include both the smectic layering (via the layer displacement $u$ and the layer normal $\hat{\rho}$) and the director $\hat{n}$ of the underlying nematic order in our macroscopic hydrodynamic description and allow both directions to differ in non equilibrium situations. In a homeotropically aligned sample the nematic director couples to an applied simple shear, whereas the smectic layering stays unchanged. This difference leads to a finite (but usually small) angle between $\hat{n}$ and $\hat{\rho}$, which we find to be equivalent to an effective dilatation of the layers. This effective dilatation leads, above a certain threshold, to an undulation instability of the layers with a wave vector parallel to the vorticity direction of the shear flow. We include the couplings of the velocity field with the order parameters for orientational and positional order and show how the order parameters interact with the undulation instability. We explore the influence of the magnitude of various material parameters on the instability. Comparing our results to available experimental results and molecular dynamic simulations, we find good qualitative agreement for the first instability. In addition, we discuss pathways to higher instabilities leading to the formation of onions (multilamellar vesicles) via cylindrical structures and/or the break-up of layers via large amplitude undulations.
When many complex fluids are submitted to an applied shear flow, they show interesting coupling phenomena between their internal structure and the flow field. For layered systems of smectic A type (including block copolymers, lyotropic systems, and side-chain liquid crystalline polymers) this coupling can induce a reorientation of the layers. Experiments on a large number of systems which differ significantly in their microscopic details nevertheless show striking similarities in their macroscopic behavior under shear. The systems under investigation include block copolymers [1–9], low molecular weight (LMW) liquid crystals [10–12], lyotropic lamellar phases (both LMW [13–21] and polymeric [22]), and liquid crystalline side-chain polymers [23–25]. The experiments performed make either use of steady shear (typically for the low viscosity systems, e.g., in a Couette cell) or large amplitude oscillatory shear (LAOS, often in the highly viscous polymeric systems, e.g., in a cone–plate or plate–plate geometry). Due to these experimental differences a direct comparison between the different systems is not always straightforward. The most common features of all these experiments can be summarized as follows. Starting with an aligned sample where the layers are parallel to the planes of constant velocity (“parallel” orientation), the layering is stable up to a certain critical shear rate [2, 5, 11–13, 17, 22]. At higher shear rates two different situations prevail. Depending on the system either multi-lamellar vesicles [13, 19–22] (“onions”, typically in lyotropic systems) or layers perpendicular to the vorticity direction [1–5, 11, 12, 24] (“perpendicular” orientation, typically in solvent free systems) are
observed. In a number of systems a third regime is observed at even higher shear rates with a parallel orientation [5, 13]. If the starting point is rather a randomly distributed lamellar phase, the first regime is not observed [1, 4, 19, 26]. This last point illustrates that experiments on layered liquids depend on the pre-history of the sample. In our further discussion we will concentrate on systems showing a well aligned parallel orientation before shear is applied.

The experimental similarities between different systems suggest that the theoretical description of these reorientations can be constructed, at least to some extent, from a common basis independent of the actual system. (On the other hand, a description including the differences between the systems under investigation must refer more closely to their microscopic details. In particular single molecule effects are expected to influence significantly the phenomena observed for polymers and block-copolymers). When looking for a macroscopic description, the well established standard smectic A hydrodynamics [27–30] might appear as a good starting point for such a theoretical approach. However, using the usual set of hydrodynamic equations of the smectic A phase [28–30] the observed change of orientation cannot be explained, because in these models each layer is assumed to be a two dimensional isotropic fluid. Or to put it differently: no destabilizing mechanism for well aligned parallel layers is present in the standard smectic A hydrodynamics.

In the framework of irreversible thermodynamics (compare, for example, [31, 32]) the macroscopic variables of a system can be divided into those due to conservation laws (here mass density \( \rho \), momentum density \( g = \rho v \) with the velocity field \( v \) and energy density \( \varepsilon \)) and those reflecting a spontaneously broken continuous symmetry (here the layer displacement \( u \) characterizes the broken translational symmetry parallel to the layer normal). For a smectic A liquid crystal the director \( \hat{n} \) of the underlying nematic order is assumed to be parallel to the layer normal \( \hat{p} \). So far, only in the vicinity of a nematic–smectic A phase transition has a finite angle between \( \hat{n} \) and \( \hat{p} \) been shown to be of physical interest [33].

Smectic A liquid crystals are known to be rather sensitive to dilatations of the layers. As shown in [34, 35], a relative dilatation of less than \( 10^{-4} \) parallel to the layer normal suffices to cause an undulation instability of the smectic layers. Above this very small, but finite, critical dilatation the liquid crystal develops undulations of the layers to reduce the strain locally. Later on Oswald and Ben-Abraham considered dilated smectic A under shear [36]. When a shear flow is applied (with a parallel orientation of the layers), the onset for undulations is unchanged only if the wave vector of the undulations points in the vorticity direction (a similar situation was later considered in [37]). Whenever this wave vector has a component in the flow direction, the onset of the undulation instability is increased by a portion proportional to the applied shear rate.

Over the last decade several explanations have been proposed for specific systems. In [38] the effect of shear flow on layer fluctuations in lamellar phases has been considered. The authors found that the lifetime of thermal fluctuations is significantly influenced by the shear flow and concluded that this can give rise to a destabilization of the layers. In [39], Williams and MacKintosh calculated the effect of the tangential strain on each layer in a sheared block copolymer. By minimization
of the free energy of the system they found a tilt of the polymer chains and a tendency of the layers to reduce their thickness. They interpreted this tendency as a dilatation and found an undulation instability by similar arguments as those given in [34, 35]. About a decade ago Zilman and Granek [40] considered short wavelength fluctuations. In their model these fluctuations are suppressed for energetic reasons leading to an undulation instability of the layers. More recently Marlow and Olmsted investigated the importance of the entropic Helfrich interactions [41] for lyotropic systems and found the possibility of an undulational instability for non-permeable or only slowly permeating membranes.

Throughout this chapter we focus on the extended hydrodynamic description for smectic A-type systems presented in [42, 43]. We discuss the possibility of an undulation instability of the layers under shear flow keeping the layer thickness and the total number of layers constant. In contrast to previous approaches, Auernhammer et al. derived the set of macroscopic dynamic equations within the framework of irreversible thermodynamics (which allows the inclusion of dissipative as well as reversible effects) and performed a linear stability analysis of these equations. The key point in this model is to take into account both the layer displacement $u$ and the director field $\hat{n}$. The director $\hat{n}$ is coupled elastically to the layer normal $\hat{p} = \frac{\nabla (z-u)}{|\nabla (z-u)|}$ in such a way that $\hat{n}$ and $\hat{p}$ are parallel in equilibrium; $z$ is the coordinate perpendicular to the plates.

This chapter is organized as follows. After a review of the model presenting the macroscopic equations in Sect. 2.1 and their implementation in Sect. 2.2, we extend the basic model in the following sections. Especially we include the cross coupling to the velocity field and the moduli of the nematic and smectic order parameters. It turns out that the coupling terms to the velocity are important since they can change the critical parameters significantly. We find that the moduli of the order parameters also show undulations and, thus, regions with a reduced order parameter can be identified. The comparison of the different levels of approximations shows that the basic model is contained in this more general analysis as a special case. We also compare our results to experiments and molecular dynamic simulations and show that an oscillatory instability is extremely unlikely to occur. In the last two sections we discuss higher instabilities leading to the formation of onions (multilamellar vesicles, MLVs) via cylindrical structures and/or the break-up of layers due to large amplitude undulations, pathways that have also been suggested in scattering experiments, in particular in Richtering’s group.

2 Model and Technique

In this and the next section we will draw heavily on [43] and we will also make use of our older work [42].
2.1 Physical Idea of the Model

In a smectic A liquid crystal one can easily define two directions: the normal to the layers \( \hat{p} \) and an average over the molecular axes, the director, \( \hat{n} \). In the standard formulation of smectic A hydrodynamics these two directions are parallel by construction. Only in the vicinity of phase transitions (either the nematic–smectic A or smectic A–smectic C*) has it been shown that director fluctuations are of physical interest [33, 44, 45]. Nevertheless \( \hat{n} \) and \( \hat{p} \) differ significantly in their interaction with an applied shear flow.

We consider a situation as show in Fig. 1. A well aligned smectic A liquid crystal is placed between two parallel and laterally infinite plates. The upper plate (located at \( z = d/2 \)) moves with a constant velocity \( v_u = d\dot{\gamma}\hat{e}_x/2 \) to the right and the lower plate (at \( -d/2 \)) moves with the same velocity in the opposite direction \( (v_l = -d\dot{\gamma}\hat{e}_x/2) \). Thus the sample is submitted to an average shear given by \( (v_u - v_l)/d = \dot{\gamma} \). As mentioned above, a three dimensional stack of parallel fluid layers cannot couple directly to an applied shear flow. Neither does the layer normal: it stays unchanged as long as the flow direction lies within the layers. In contrast, it is
well known from nematodynamics that the director experiences a torque in a shear flow. This torque leads – in the simplest case – to a flow aligning behavior of the director. The key assumption in the model of [42] is that this torque is still present in a smectic A liquid crystal and acts only on the director \( \hat{n} \) and not on the layer normal \( \hat{p} \). An energetic coupling between \( \hat{n} \) and \( \hat{p} \) ensures that both directions are parallel in equilibrium.

Submitted to a shear flow the layer normal will stay unchanged, but the director will tilt in the direction of the flow until the torques due to the flow and due to the coupling to the layer normal balance one another. For any given shear rate a finite, but usually small, angle \( \theta \) between \( \hat{n} \) and \( \hat{p} \) will result. This finite angle has important consequences for the layers: Since the preferred thickness of the layers is proportional to the projection of the director on the layer normal, a finite angle between those two directions is equivalent to an effective dilatation of approximately \( \theta^2/2 \) (see Fig. 2). If we assume a constant total sample thickness and exclude effects of defects, the system can accommodate this constraint by layer rotations. A global rotation of the layers is not possible, but they can rotate locally (as in the case of dilated smectic A liquid crystals [34, 35]). This local rotation of the layers leads to undulations as shown in Fig. 3. These undulation are a compromise between the effective dilatation (which is not favorable for the system) and the curvature of the layers due to the undulations (which costs energy). In the static case of dilated smectic A liquid crystals no direction is preferred, but Oswald and Ben-Abraham [36] have shown that this symmetry is broken if an additional shear is applied to the system. In this case the standard formulation of smectic A hydrodynamics predicts that the wave vector of the undulations will point along the neutral direction of the shear (vorticity direction). In this chapter we will assume that this result of Oswald and Ben-Abraham also holds in the case of our extended formulation of smectic A hydrodynamics (see Fig. 3).

Fig. 2 A finite angle \( \theta \) between \( \hat{n} \) and \( \hat{p} \) leads to a tendency of the layers to reduce their thickness. Assuming a constant number of layers in the sample, this tendency is equivalent to an effective dilatation of the layers. For small angles between \( \hat{n} \) and \( \hat{p} \) the relative effective dilatation is given by \( \theta^2/2 \).
2.2 Implementation of the Model

To generate the macroscopic hydrodynamic equations we follow the procedure given by the framework of irreversible thermodynamics [31]. This method has successfully been applied in many cases to derive the macroscopic hydrodynamic equations of complex fluids (see, e.g., [28, 30, 42, 46, 47]). The advantage of this method is its systematic way of deducing the governing equations. Once the set of variables is given, the macroscopic hydrodynamic equations follow by applying basic symmetry arguments and thermodynamic considerations.

Let us briefly review the essential ingredients to this procedure (for more details of the method see [30] and for our model [42]). For a given system the hydrodynamic variables can be split up into two categories: variables reflecting conserved quantities (e.g., the linear momentum density, the mass density, etc.) and variables due to spontaneously broken continuous symmetries (e.g., the nematic director or the layer displacements of the smectic layers). Additionally, in some cases non-hydrodynamic variables (e.g., the strength of the order parameter [48]) can show slow dynamics which can be described within this framework (see, e.g., [30, 47]).

Using all these variables the relations, which form the starting point for the further calculations, can be constructed. These relations are: the energy density $\varepsilon$, the dissipation function $R$, the Gibbs-relation and the Gibbs–Duhem relation. To illustrate the idea of our model we split up $\varepsilon$ and $R$ into several parts according to the different origin of the variables:

- Conserved quantities (index cons)
- Symmetry variables (index sym)
- The modulus of the order parameter (index ord)

In the spirit of our model, two order parameters play a role: the nematic tensorial order parameter $Q_{ij}$ and the smectic $A$ complex order parameter $\Phi$. For practical reason we use the director $\hat{n}$ and the modulus $S^{(n)}$ in the uniaxial nematic case.
\[ Q_{ij} = \frac{3}{2} S^{(n)} (n_i n_j - \frac{1}{3} \delta_{ij}) \] and the layer displacement \( u \) and the modulus \( S^{(s)} \) in the smectic A case \( \varphi = S^{(s)} \exp \{ i q_0 (z - u) \} \). Here, as in the rest of the chapter, we refer to the system of coordinates defined in Sect. 2.1. We note that \( u \) is only a good variable if we consider small deformations of the layers. For large layer deformations the phase \( \varphi = z - u \) is the appropriate variable \([49, 50]\). In our further discussion, we will concentrate on the parts due to symmetry variables and the order parameters, while for terms already present in the isotropic fluid see, e.g., \([30, 31]\).

Let us first consider the energy density. The conventions of notation introduced by the following equations are summarized in Table 1:

\[ \varepsilon = \varepsilon_{\text{cons}} + \varepsilon_{\text{sym}} + \varepsilon_{\text{ord}}^{(n)} + \varepsilon_{\text{ord}}^{(s)} \] (1)

\( \varepsilon_{\text{cons}} \), which is identical to the isotropic fluid, is discussed elsewhere \([30, 31]\). The symmetry part reads

\[ \varepsilon_{\text{sym}} = \frac{1}{2} K_1 (\nabla \cdot \hat{n})^2 + \frac{1}{2} K_2 [\hat{n} \cdot (\nabla \times \hat{n})]^2 + \frac{1}{2} K_3 [(\nabla \times \hat{n})]^2 + \frac{1}{2} K (\nabla_{\perp} u)^2 \]

\[ + \frac{1}{2} B_0 \left[ \nabla_z u + (1 - n_z) - \frac{1}{2} (\nabla_{\perp} u)^2 \right]^2 + \frac{1}{2} B_1 (\hat{n} \times \hat{p})^2 \] (2)

In (2) the spirit of the model becomes clear. We combine the properties of a nematic liquid crystal (the first two lines) with those of a smectic \( A \) (the third and fourth lines) and couple both parts (the last lines) in such a way that \( \hat{n} \) and \( \hat{p} \) are parallel.

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Explicit form</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>( K )</td>
<td>( K )</td>
<td>Bending modulus of layers</td>
</tr>
<tr>
<td>( B_0 )</td>
<td>( B_0 )</td>
<td>Compressibility of layers</td>
</tr>
<tr>
<td>( B_1 )</td>
<td>( B_1 )</td>
<td>Coupling between the director and the layer normal</td>
</tr>
<tr>
<td>( L_0^{(n,s)} )</td>
<td>( L_0^{(n,s)} )</td>
<td>Variations of the order parameter (nematic and smectic, respectively)</td>
</tr>
<tr>
<td>( L_{1,ij}^{(n)} )</td>
<td>( L_{1,ij}^{(n)} \delta_{ij} + L_{1,ij}^{(n)} n_i n_j )</td>
<td>Gradients terms of the order parameter (nematic)</td>
</tr>
<tr>
<td>( M_{ijk} )</td>
<td>( M_0 (\delta_{ij} n_k + \delta_{ik} n_j) )</td>
<td>Cross-coupling between the director and order parameter (nematic)</td>
</tr>
<tr>
<td>( L_{1,ij}^{(s)} )</td>
<td>( L_{1,ij}^{(s)} (\delta_{ij} - p_i p_j) + L_{1,ij}^{(s)} p_i p_j )</td>
<td>Gradients terms of the order parameter (smectic)</td>
</tr>
</tbody>
</table>
in equilibrium. As discussed earlier [42], $\varepsilon_{\text{sym}}$ simplifies considerably by dropping higher order terms and assuming a small angle between $\hat{n}$ and $\hat{p}$. Splay deformations of the director are generally considered as higher order corrections to dilatations of the smectic layers. Twist deformations are forbidden in standard smectic A hydrodynamics and must be small as long as the angle between $\hat{n}$ and $\hat{p}$ is small. Additionally, the difference between the splay deformation of the director field $K_1/2 (\nabla \cdot \hat{n})^2$ and bending of the layers $K/2 (\nabla u)^2$ is negligible. Consequently we combine splay and bend in a single term with a single elastic constant which we call $K'$: $K_1/2 (\nabla \cdot \hat{n})^2 + K/2 (\nabla u)^2 \approx K'/2 (\nabla u)^2$. In the following we drop the prime and call the new elastic constant $K$. The approximated version of $\varepsilon_{\text{sym}}$ is now given by

$$
\varepsilon_{\text{sym}} = \frac{1}{2} K (\nabla u)^2 + \frac{1}{2} B_0 \left[ \nabla u + \left( 1 - n_z \right) - \frac{1}{2} (\nabla u)^2 \right]^2 + \frac{1}{2} B_1 \left( \hat{n} \times \hat{p} \right)^2
$$

(3)

In our model the moduli of the nematic and smectic order parameters play similar roles, so we will deal with both. Since we consider a situation beyond the phase transition regime, the equilibrium value of the order parameter is non-zero ($S_{0(n,s)}$, for both nematic and smectic) and only its variations $s^{(n,s)}$ can enter the energy density ($S^{(n,s)} = S_{0(n,s)} + s^{(n,s)}$):

$$
\varepsilon^{(n)}_{\text{ord}} = \frac{1}{2} L_0 \left( s^{(n)} \right)^2 + \frac{1}{2} L_{1,ij}^{(n)} \left( \nabla_i s^{(n)} \right) \left( \nabla_j s^{(n)} \right) + M_{ijk} \nabla_j n_i \nabla_k s^{(n)}
$$

(4)

$$
\varepsilon^{(s)}_{\text{ord}} = \frac{1}{2} L_0 \left( s^{(s)} \right)^2 + \frac{1}{2} L_{1,ij}^{(s)} \left( \nabla_i s^{(s)} \right) \left( \nabla_j s^{(s)} \right)
$$

(5)

By a similar construction we write down the dissipation function as (see Table 2 for a list of the thermodynamic variables and their conjugates):

**Table 2** Variables and their conjugates, i.e., the corresponding thermodynamic forces

<table>
<thead>
<tr>
<th>Name</th>
<th>Variable</th>
<th>Conjugate</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass density</td>
<td>$\rho$</td>
<td>$\mu$</td>
</tr>
<tr>
<td>Momentum density</td>
<td>$g$</td>
<td>$v$</td>
</tr>
<tr>
<td>Nematic director</td>
<td>$\hat{n}$</td>
<td>$\hat{h}$</td>
</tr>
<tr>
<td>Smectic layer displacement</td>
<td>$u$</td>
<td>$\Psi$</td>
</tr>
<tr>
<td>Variation of the modulus of the order parameter (either nematic or smectic)</td>
<td>$s^{(n,s)}$</td>
<td>$\Xi^{(n,s)}$</td>
</tr>
</tbody>
</table>
\[ R = R_{\text{cons}} + R_{\text{sym}} + R_{\text{ord}} \quad (6) \]

\[ R_{\text{cons}} = \frac{1}{2} \nu_{ijkl} (\nabla_j v_i) (\nabla_l v_k) + R_0 \quad (7) \]

\[ R_{\text{sym}} = \frac{1}{2 \gamma} h_i \delta_{ij} h_j + \lambda_p \psi^2 \quad (8) \]

\[ R_{\text{ord}} = \frac{1}{2} \alpha^{(n)} (\Xi^{(n)})^2 + \frac{1}{2} \alpha^{(s)} (\Xi^{(s)})^2 \quad (9) \]

where \( R_0 \) summarizes further terms due to conservations laws, which do not enter our further calculation, and (after [46])

\[ \nu_{ijkl} = \nu_2 (\delta_{jl} \delta_{ik} + \delta_{il} \delta_{jk}) \]

\[ + 2 (\nu_1 + \nu_2 - 2 \nu_3) n_i n_j n_k n_l \]

\[ + (\nu_3 - \nu_2) (n_i n_l \delta_{jk} + n_j n_k \delta_{il}) \]

\[ + n_i n_k \delta_{jl} + n_i n_l \delta_{jk}) \]

\[ + (\nu_4 - \nu_2) \delta_{ij} \delta_{kl} \]

\[ + (\nu_5 - \nu_4 + \nu_2) (\delta_{j} n_k n_l + \delta_{kl} n_i n_j) \quad (10) \]

As mentioned in Sect. 2.1, we consider a shear induced smectic C like situation (but with a small tilt angle, i.e., a weak biaxiality). We neglect this weak biaxiality in the viscosity tensor and use it in the uniaxial formulation given above (with the director \( \hat{n} \) as the preferred direction). This assumption is justified by the fact that the results presented in this chapter do not change significantly if we use \( \hat{p} \) instead of \( \hat{n} \) in the viscosity tensor.

Throughout our calculations we will not assume any restriction on the viscosity constants except the usual requirements due to thermodynamic stability (see, e.g., [30]). Later on we will impose the incompressibility of the fluid by assuming a constant mass density \( \rho \) of the fluid. We emphasize that this procedure does not require any further assumption about the material parameters.

The set of basic equations is completed by the Gibbs–Duhem (the local formulation of the second law of thermodynamics) and the Gibbs relation (which connects the pressure \( P \) with the other thermodynamic quantities), which we will use in the following form:

\[ d\epsilon = d\epsilon_0 + v d\gamma + \phi_{ij} d\nabla_j n_i + h_i' d n_i + \psi_i d\nabla_i u \]

\[ + \Xi^{(n)} d s^{(n)} + \Xi_i^{(n)} d\nabla_i s^{(n)} \]

\[ + \Xi^{(s)} d s^{(s)} + \Xi_i^{(s)} d\nabla_i s^{(s)} \quad (11) \]
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\[ P = -\varepsilon + \mu \rho + T \sigma + v \cdot g \]  \hspace{1cm} (12)

The newly defined quantities in (11) are connected to the thermodynamic forces (Table 2) by the following relations:

\[ h_i = h_i' - \nabla_j \phi_{ij} = \frac{\delta \varepsilon}{\delta n_i} \]  \hspace{1cm} (13)

\[ \Psi = -\nabla_i \psi_i = \frac{\delta \varepsilon}{\delta u} \]  \hspace{1cm} (14)

\[ \Xi^{(n,s)} = \Xi'^{(n,s)} - \nabla_i \Xi''^{(n,s)} = \frac{\delta \varepsilon}{\delta \delta^{(n,s)}} \]  \hspace{1cm} (15)

Following the standard procedure within the framework of irreversible thermodynamics we find the following set of macroscopic hydrodynamic equations [30,31,42,47]:

\[ \frac{\partial}{\partial t} u + v_j \nabla_j u = v_z - \lambda_p \Psi \]  \hspace{1cm} (16)

\[ \frac{\partial}{\partial t} n_i + v_j \nabla_j n_i = \frac{1}{2} \left[ (\lambda - 1) \delta_{ij} n_k + (\lambda + 1) \delta_{ik} n_j \right] \nabla_j v_k - \frac{1}{\gamma} \delta_{ik} h_k \]  \hspace{1cm} (17)

\[ 0 = \nabla_i v_i \]  \hspace{1cm} (18)

\[ \rho \left( \frac{\partial}{\partial t} v_i + v_j \nabla_j v_i \right) = -\nabla_j \left\{ \psi_j (\nabla_i u + \delta_{i3}) + \beta^{(n,s)}_{ij} \Xi^{(n,s)} \right\} \\
- \frac{1}{2} \left[ (\lambda - 1) \delta_{jk} n_i + (\lambda + 1) \delta_{ik} n_j \right] h_k \\
+ v_{ijkl} \nabla_l v_k \right\} - \nabla_i P \]  \hspace{1cm} (19)

\[ \frac{\partial}{\partial t} \delta^{(n,s)} + v_j \nabla_j \delta^{(n,s)} \]  \hspace{1cm} (20)
For the reversible parts of the equations some coupling constants have been introduced: the flow-alignment tensor

$$\lambda_{ijk} = \frac{1}{2} \left[ (\lambda - 1) \delta^\top_{ij} n_k + (\lambda + 1) \delta^\parallel_{ik} n_j \right]$$  \hspace{1cm} (21)

with the flow-alignment parameter $\lambda$ (and using $\delta^\top_{ij} = \delta_{ij} - n_i n_j$) and the coupling between flow and order parameter

$$\beta^{(n)}_{ij} = \beta^{(n)}_\perp \delta^\perp_{ij} + \beta^{(n)}_\parallel n_i n_j$$  \hspace{1cm} (22)

$$\beta^{(s)}_{ij} = \beta^{(s)}_\perp (\delta_{ij} - p_i p_j) + \beta^{(s)}_\parallel p_i p_j$$  \hspace{1cm} (23)

Furthermore, there is a reversible coupling between the layer displacement and the velocity field in (16). But its coupling constant has to be unity due to the Gallilei invariance of the equations. As mentioned above, the use of $u$ is limited to small layer deformations.

The transverse Kronecker symbols $\delta^\perp_{ij}$ in (17) and (21) guarantee the normalization of $\hat{n}$. This implies that only two of the (17) are independent. For the following calculations it turned out to be useful to guarantee the normalization of the director by introducing two angular variables $\theta$ and $\phi$ to describe the director:

$$n_x = \sin \theta \cos \phi$$  \hspace{1cm} (24)

$$n_y = \sin \theta \sin \phi$$  \hspace{1cm} (25)

$$n_z = \cos \theta$$  \hspace{1cm} (26)

Consequently, (17) has to be replaced using angular variables. Denoting the right hand side of (17) with $Y_i$, this can be done the following way:

$$\frac{\partial}{\partial t} \theta + v_j \nabla_j \theta = Y_x \cos \theta \cos \phi + Y_z \cos \theta \sin \phi - Y_z \sin \theta$$  \hspace{1cm} (27)

$$\frac{\partial}{\partial t} \phi + v_j \nabla_j \phi = -Y_x \sin \phi \sin \theta + Y_z \cos \phi \sin \theta$$  \hspace{1cm} (28)

In the same way, we guarantee the normalization of $\hat{p}$ by using

$$p_x = 0$$  \hspace{1cm} (29)

$$p_y = -\nabla_y u$$  \hspace{1cm} (30)

$$p_z = \sqrt{1 - p_y^2}$$  \hspace{1cm} (31)

The different ways of normalizing $\hat{n}$ and $\hat{p}$ arise from the fact that $\hat{p}$ is parallel to $\hat{e}_z$ in zeroth order, whereas $\hat{n}$ encloses a finite angle with $\hat{e}_z$ for any given shear rate.
The set of macroscopic hydrodynamic equations we now deal with, (16), (18)–(20), (27), and (28), follows directly from the initial input in the energy density and the dissipation function without any further assumptions.

To solve these equations we need suitable boundary conditions. In the following we will assume that the boundaries have no orienting effect on the director (the homeotropic alignment of the director is only due to the layering and the coupling between the layer normal $\hat{p}$ and the director $\hat{n}$). Any variation of the layer displacement must vanish at the boundaries:

$$u \left( \pm \frac{1}{2} d \right) = 0$$  \hspace{1cm} (32)

For the velocity field the situation is a little more complex: We assume no-slip boundary conditions, i.e., the velocity of the fluid and the velocity of the plate are the same at the surface of the plate. It is convenient to split the velocity field into two parts: the shear field $v_0$ which satisfies the governing equations and the no-slip boundary condition and the correction $v_1$ to this shear field. The boundary condition for $v_1$ now reads

$$v_1 \left( \pm \frac{1}{2} d \right) = 0$$  \hspace{1cm} (33)

Making use of the following considerations this condition can be simplified. Due to (16) the $z$-component of $v_1$ is suppressed by a factor of $\lambda_p$ (which is typically extremely small [29, 36]). Making use of the results of [36] we can assume that $v_1$ depends only on $y$ and $z$ and thus conclude [with (18)] that the $y$-component of $v_1$ is also suppressed by $\lambda_p$. For this reason one can assume that $v_{1,y}$ and $v_{1,z}$ are negligible and the only relevant boundary condition for the velocity field is

$$v_{1,x} = 0$$  \hspace{1cm} (34)

The validity of this assumption is nicely illustrated by our results. Figure 7 shows that $v_y$ and $v_z$ are indeed suppressed by $\lambda_p$.

### 2.3 Technique of Solution

The aim now is twofold: finding a spatially homogeneous solution of the governing equations (for a given shear rate) and investigating the stability of this solution. In this section we will describe the general procedure and give the results in Sect. 3.

We write the solution as the vector $X = (\theta, \phi, u, v_x, v_y, v_z, P, s^{(n,s)})$ consisting of the angular variables of the director, the layer displacement, the velocity field, the pressure, and the modulus of the (nematic or smectic) order parameter. For a spatially homogeneous situation the equations simplify significantly and the desired solution $X_0$ can directly be found (see Sect. 3.1). To determine the region of stability of $X_0$ we perform a linear stability analysis, i.e., we add a small perturbation $X_1$ to
If the symmetry under inversion of $z$ is given for one component of $X_1$, the symmetry of all other components follows directly from the linearized set of equations. Here we give the $z$-symmetry of all components assuming that $u$ is an even function of $z$.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>$z$-Symmetry</th>
<th>Quantity</th>
<th>$z$-Symmetry</th>
</tr>
</thead>
<tbody>
<tr>
<td>$u$</td>
<td>Even</td>
<td>$v_x$</td>
<td>Even</td>
</tr>
<tr>
<td>$\theta$</td>
<td>Odd</td>
<td>$v_y$</td>
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<tr>
<td>$\phi$</td>
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<td>$v_z$</td>
<td>Even</td>
</tr>
<tr>
<td>$P$</td>
<td>Odd</td>
<td>$s^{(n,x)}$</td>
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</table>

The homogeneous solutions $X_0: X = X_0 + X_1$ (with $X_1 \ll X_0$) and linearize the governing equations in the small perturbations. In short, the solution of the equation $L X_1 = \frac{\partial}{\partial t} X_1$ is analyzed. Here $L$ denotes the operator for the linearized set of the governing equations. The ansatz for the unknown quantities must fulfill the boundary conditions [see the discussion following (32)] and follow the symmetry scheme given by Table 3. Assuming an exponential time dependence and harmonic spatial dependence of $X_1$:

$$X_{1,i} \sim \exp \left[ (i\omega + \frac{1}{\tau} t) \right] \begin{cases} \cos(qy) \\ \sin(qy) \end{cases} \begin{cases} \cos(qz) \\ \sin(qz) \end{cases}$$

fulfills all requirements (with an oscillation rate $\omega$, a growth rate $1/\tau$, and a wave vector $q = q_x \hat{e}_x + q_z \hat{e}_z$). In this ansatz we made use of the results by Oswald and Ben-Abraham [36], who have shown that in standard dilated smectic A under shear the first instability will set in with a wave vector along the neutral direction of the flow ($q \cdot \hat{e}_x = 0$). After inserting the above ansatz in the linearized set of (partial differential) equations, a set of coupled linear equations is obtained to determine $1/\tau \text{ and } \omega$.

From the standard smectic A hydrodynamics it is known that shear does not destabilize the layers. Since our extended formulation of the smectic A hydrodynamics is equivalent to the standard smectic A hydrodynamics for vanishing external fields (e.g., shear rate), we assume that the layers are stable for low enough shear rates, i.e., $1/\tau < 0$ for small shear rates. So $1/\tau = 0$ marks the set of external parameters (shear rate) and material parameters above which $X_1$ grows. Typically we hold the material parameters fixed and the only external parameter is the shear rate. The solvability condition of the corresponding set of linear equations gives a relation between the shear rate [and tilt angle $\theta_0$, which is directly connected to the shear rate, see (38) below], $\omega$ and the wave vector $q$. For every given $q$ a specific shear rate (and tilt angle $\theta_0$) can be determined which separates the stable region (below) from the unstable region (above). This defines the so-called curve of marginal stability (or neutral curve) $\theta_0(q)$. If, for any given set of external parameters, the tilt angle $\theta_0$ lies above the curve of marginal stability for at least one value of $q$, the spatial homogeneous state is unstable and undulations grow. The smallest shear rate (tilt angle) for which undulations can grow is called the critical shear rate (tilt angle). Technically
speaking, we solve $LX = i\omega X$ in many cases we can set $\omega = 0$, see below. We point out that this linear analysis is only valid at the point where the first instability sets in. Without further investigations no prediction of the spatial structure of the developing instability can be made. Also the nature of the bifurcation (backward or forward) must be determined by further investigations.

For practical reasons we used dimensionless units in our numerical calculations. The invariance of the governing equations under rescaling time, length, and mass allows us to choose three parameters in these equations to be equal to unity. We will set

$$B_1 = 1, \quad \gamma_1 = 1, \quad \text{and} \quad \frac{d}{\pi} = q_z = 1 \quad (36)$$

and measure all other quantities in the units defined by this choice. Nevertheless we will keep these quantities explicitly in our analytical work.

To extract concrete predictions for experimental parameters from our calculations is a non-trivial task, because neither the energetic constant $B_1$ nor the rotational viscosity $\gamma_1$ are used for the hydrodynamic description of the smectic A phase (but play an important role in our model). Therefore, we rely here on measurements in the vicinity of the nematic–smectic A phase transition. Measurements on LMW liquid crystals made by Litster [33] in the vicinity of the nematic–smectic A transition indicate that $B_1$ is approximately one order of magnitude less than $B_0$. As for $\gamma_1$ we could not find any measurements which would allow an estimate of its value in the smectic A phase. In the nematic phase $\gamma_1$ increases drastically towards the nematic–smectic A transition (see, e.g., [51]). Numerical simulations on a molecular scale are also a promising approach to determine these constants [52].

Due to the complexity of the full set of governing equations, we will start our analysis with a minimal set of variables ($\theta$, $\phi$, and $u$) and suppress the coupling to the other variables (see Sect. 3.2.1). Step by step the other variables will be taken into account. The general picture of the instability will turn out to be already present in the minimal model, but many interesting details will be added throughout the next sections. In comparison to our earlier work [42], we now use the way of normalizing $\hat{n}$ and $\hat{p}$ derived above. This will lead to some small differences in the results but leaves the general picture unchanged. First we assume a stationary instability (i.e., we let $\omega = 0$); later on we discuss the possibility for an oscillatory instability and have a look at some special features of the system (Sects. 3.3 and 3.4).

3 Results and Discussion

3.1 Spatially Homogeneous State

Looking for a spatially homogeneous solution, the governing equations simplify significantly. A linear shear profile

$$v_0 = \gamma z \hat{e}_x \quad (37)$$
is a solution to (18) and $u$ stays unchanged in this regime. The only variables which have a zeroth order correction for all shear rates are the tilt angle $\theta$ and the modulus of the nematic order parameters $s^{(n)}$:

$$
\left( \frac{\lambda + 1}{2} - \lambda \sin^2(\theta_0) \right) \dot{\gamma} = \frac{B_1}{\gamma_1} \sin(\theta_0) \cos(\theta_0) + \frac{B_0}{\gamma_1} \sin(\theta_0)(1 - \cos(\theta_0))
$$

(38)

Equation (39) shows that nematic degrees of freedom couple to simple shear, but not the smectic degrees of freedom; the modulus of the nematic order parameter has a non-vanishing spatially homogeneous correction (see (39)), whereas the smectic order parameter stays unchanged. The reason for this difference lies in the fact that $\beta_{\parallel}^{(n)}$ and $\beta_{\perp}^{(n)}$ include $\hat{n}$ and $\hat{p}$, respectively, which coupled differently to the flow field (see (22) and (23)). Equation (38) gives a well defined relation between the shear rate $\dot{\gamma}$ and the director tilt angle $\theta_0$, which we will use to eliminate $\dot{\gamma}$ from our further calculations. To lowest order $\theta_0$ depends linearly on $\dot{\gamma}$:

$$
\theta_0 = \frac{\dot{\gamma}}{B_1} \frac{\lambda + 1}{2} + O(\theta_0^3)
$$

(40)

We are not aware of any experimental data, which would allow a direct comparison with these results. We stress, however, that molecular dynamics simulations by Soddemann et al. [53] are in very good agreement with (38) and (40).

In contrast to the director tilt the lowest order correction to the nematic order parameter is quadratic in the shear rate (tilt angle):

$$
s_0^{(n)} = \frac{2}{\lambda + 1} \frac{B_1}{\gamma_1} \frac{\beta_{\parallel}^{(n)} - \beta_{\perp}^{(n)}}{\alpha^{(n)} L_0} \theta_0^2 + O(\theta_0^4)
$$

(41)

In the following we consider perturbations around the spatially homogeneous state given above.

### 3.2 Stationary Instability

#### 3.2.1 Minimal Set of Variables

Let us first consider the effect of our modifications regarding the normalization of $\hat{n}$ and $\hat{p}$ in comparison to our earlier results [42]. For this purpose we consider only
a minimal set of variables: the director (characterized by the two angles $\theta$ and $\phi$) and the layer displacement $u$. We neglect all couplings of these variables to other quantities describing the system, namely the velocity field and the moduli of the nematic and smectic order parameters. Within these approximations the equations to solve are

\begin{align}
0 &= A_\theta \left\{ 2\gamma \lambda \sin(\theta_0) \cos(\theta_0) \\
&+ \frac{B_0}{\gamma_1} \left[ \sin^2(\theta_0) - \cos^2(\theta_0) + \cos(\theta_0) \right] \\
&- \frac{B_1}{\gamma_1} \left[ \sin^2(\theta_0) - \cos^2(\theta_0) \right] \right\} \\
&- A_u \frac{B_0}{\gamma_1} \sin(\theta_0) q_z
\end{align}  

(42)

\begin{align}
0 &= A_\phi \frac{1}{2} \gamma (\lambda + 1) - A_u \frac{B_1}{\gamma_1} q
\end{align}  

(43)

\begin{align}
0 &= A_\theta \lambda_\rho B_0 \sin(\theta_0) q_z \\
&+ A_\phi \lambda_\rho B_1 q \sin(\theta_0) \cos(\theta_0) \\
&- A_u \lambda_\rho \left[ -B_0 q^2 (1 - \cos(\theta_0)) \right. \\
&\left. + B_1 q^2 \cos^2(\theta_0) + Kq^4 + B_0 q_z^2 \right]
\end{align}  

(44)

Here we inserted an ansatz of the type (35) and denoted the linear amplitudes of $\theta$, $\phi$, and $u$ by $A_\theta$, $A_\phi$, and $A_u$, respectively. One can solve these equations either by expanding them in a power series of $\theta_0$ (expecting to get a closed result for the critical values) or numerically. It turns out that one has to take into account terms (at least) up to order $\theta_0^5$ in (42)–(44) to get physically meaningful (but rather long and complicated) analytical results. For this reason the closed expressions have no advantage over the purely numerical solutions and we do not give the analytical approximations explicitly. A comparison with the results of [42] will be given in the Appendix. We will present and discuss our findings using the minimal set of variables in Sect. 3.2.2 in direct comparison to the results of the full set of equations.

### 3.2.2 Coupling to the Velocity Field

In the previous section we have shown that a minimal set of variables supports our picture of the physical mechanism. But neglecting the coupling between velocity field and nematic director and vice versa is a rather crude approximation since it is well known, that this coupling plays an important role in nematohydrodynamics [29,30]. So the natural next step is to include this coupling and to perform a linear
stability analysis of (16)–(18), (27) and (28). In this case the standard procedures leads to a system of seven coupled linear differential equations. Following the discussion after (32) these equations can be solved by an ansatz of the type given in (35). This reduces the system of equations to seven coupled linear equations which are easily solved using standard numerical tools (such as singular value decomposition and inverse iteration to find the eigenvectors). Due to the complexity of the equations we used Maple to determine the final set of linear equations. The key ingredients of this Maple script are given in [54].

Figure 4 gives a comparison of typical neutral curves for the minimal model and calculations including the velocity field. The overall shape of the neutral curve is not changed due to the coupling to the velocity field but a shift of the critical values (especially in the critical tilt angle) is already visible. The inset shows the relative amplitudes of the linear solutions at onset on a logarithmic scale. For \( \theta, \phi, \) and \( u \) the left bars correspond to the minimal model and the right bars to the extended version. Note that amplitudes with a different sign are shown with a different line style in the histograms (see figure caption for details).

Let us have a closer look at the differences between the minimal and the extended set of equations and follow these differences along some paths in the parameter space. As mentioned in Sect. 2.3, we can omit some of the physical parameters by using dimensionless parameters. In Figs. 5–9 we show the dependence of the critical values of the tilt angle and wave vector on the dimensionless parameters (as defined

![Figure 4](image_url)

**Fig. 4** A typical picture for the comparison of the neutral curves using the minimal set of variables (solid line) and including the velocity fields (dashed line). The overall behavior does not change but the critical values are altered due to the coupling with the velocity field. For this plot we used (in the dimensionless units discussed in Sect. 2.3) \( B_0 = 10, K = 10^{-6}, \lambda = 1.1, v_1 = v_2 = v_3 = v_4 = v_5 = 0.1, \) and \( \lambda_p = 10^{-6} \). The inset shows the linear amplitudes \( A_i \) (where \( i \) stands for \( \theta, \phi, \) etc.) at onset. Since the logarithm of the amplitudes is shown, amplitudes with different sign are shown with a different line style. Using the minimal set (left bars) all amplitudes have the same sign (solid lines). Including the velocity field (right bars) some amplitudes are positive (dashed lines), others negative (dotted lines). Note that we use in this and all following plots the dimensionless units defined by (36).
Fig. 5 A significant difference between the various approaches is only visible for $B_0 \lesssim 100$. At higher values of $B_0$ the number of free variables plays no noticeable role and the critical values follow a master curve. The solid lines show results including the velocity field, the dashed lines correspond to the minimal set of variables. At low $B_0$ in the upper curves we used $\lambda = 2$

Fig. 6 Plotting the critical values as a function of the bending modulus $K$ shows a convergence of the curves, which is nevertheless not as pronounced as in the case of Fig. 5. The influence of $\lambda$ on the critical tilt angle is significant ($\lambda = 2$ in the upper curves and $\lambda = 1.1$ in the lower ones). Again the solid lines show results including the velocity field, the dashed lines correspond to the minimal set of variables, and the dotted lines depict the outcome of the first approach. Note that the wave vectors of the minimal set and of the calculations including the velocity field are indistinguishable within the resolution of the plot

in (36)). For all these figures we used the same basic set of parameters: $B_0 = 10$, $K = 10^{-6}$, $\lambda = 1.1$, $\nu_1 = \nu_2 = \nu_3 = \nu_4 = \nu_5 = 0.1$, and $\lambda_p = 10^{-6}$. These values are estimates for a typical thermotropic LMW liquid crystal, where we made use of the results of [33, 51] (as far as $B_1$ and $\gamma_1$ are concerned, see also the last paragraph in Sect. 3.2.1). For flow alignment parameters in the range $1 \lesssim \lambda \lesssim 3$ the critical values vary strongly with $\lambda$ (see Fig. 9). Therefore we discuss in addition the situation for $\lambda = 2$ to indicate the range of possible values.
In all our calculations $v_{1,x}$ is the dominating component of $v_1$. This graph demonstrates that the other components are suppressed by $\lambda_p$ (making them almost negligible).

Considering the critical values as a function of the compression modulus $B_0$ results in a rather simple situation (Fig. 5). For small values of $B_0$ a significant influence of the coupling between the director and velocity field is apparent, which also shows a strong dependence on $\lambda$. For large $B_0$ all these differences vanish and only a single curve is obtained. At this point a comparison to dilated smectic $A$ is instructive. It is well known\cite{34,35} that in dilated smectic the critical wave vector and the critical dilatation show a power law behavior as a function of $B_0$ with exponents $1/4$ and $-1/2$, respectively. In the limit of large $B_0$ we found the same exponents already in our earlier analysis\cite{42}. If we fit power laws to our results for $B_0 > 10^2$ we find the exponents equal to $\approx 0.235$ and $\approx -0.37$ for $q_c$ and $\theta_c$, respectively (note that the dilatation in our model is $\approx \frac{1}{2} \theta_c^2$). So both approaches (the minimal set of variables and the calculations including the velocity field) show, despite all similarities to the standard model of smectic $A$ and to our earlier analysis, differences in the details of the instability.

A similar, but less pronounced, situation is apparent, when plotting the critical values as a function of the bending modulus (see Fig. 6). The curves tend to converge for large $K$, but there remains a difference between the minimal set of variables and the calculations including the the velocity field. Fitting the $K$-dependence with power laws (here for $K > 10^{-4}$) only the critical wave number exhibits an exponent close to the values expected from dilated smectic $A$ ($\approx -0.26$ vs $-\frac{1}{2}$). This illustrates the fact that shearing a lamellar system is similar to dilating it but not equivalent.

In contrast to the cases discussed above, the permeation constant $\lambda_p$ has no strong influence on the critical values. For dimensionless values $\lambda_p < 10^{-6}$ the critical values do not change at all with $\lambda_p$. For large values variations within a factor of two are possible. The permeation constant is known to be very small. In our dimensionless units we expect it be of the order of $< 10^{-9}$ for LMW thermotropic liquid crystals and neglect its influence on the critical values for this reason. In Sect. 2.2 we have emphasized that the $y$- and $z$-components of the velocity field are suppressed via $\lambda_p$. These qualitative arguments are clearly confirmed by our numerical results. In all our calculations $v_{1,x}$ is the dominating component of $v_1$ and the ratio $v_{1,y}/v_{1,x}$
Fig. 8 Only the viscosities $\nu_2$ and $\nu_3$ can influence the critical parameters significantly. The upper row depicts the dependence on a isotropic variation of the viscosity. In the middle and lower row we present the variation with $\nu_2$ and $\nu_3$ setting the other viscosities to $\nu_i = 0.1$. Here the *thick solid lines* represent the minimal set of variables. For the full set of variables we have chosen four different values of $\lambda$: the *solid curves* with $\lambda = 0.7$, the *dashed curves* with $\lambda = 1.3$, the *dotted curves* with $\lambda = 2$ and the *dot-dashed curves* with $\lambda = 3.5$. Note the similarities between the curves for small (*solid*) and large $\lambda$ (*dot-dashed*) in the upper and middle row. In these regimes $\nu_2$ is the dominating viscosity.
is of the order of $\lambda_p$ over the whole range of physical relevant values of $\lambda_p$ (see Fig. 7). This fact nicely supports our argument that we can neglect the boundary condition for $v_{1,y}$, because $v_{1,y}$ vanishes anyway.

Out of the five viscosities, only two ($\nu_2$ and $\nu_3$) show a significant influence on the critical values. In Fig. 8 we present the dependence of $\theta_c$ and $q_c$ on an assumed isotropic viscosity (upper row) and on these two viscosity coefficients (middle and lower row). Since the flow alignment parameter $\lambda$ has a remarkable influence on these curves we have chosen four different values of $\lambda$ in this figure, namely $\lambda = 0.7$, $\lambda = 1.1$, $\lambda = 2$, and $\lambda = 3.5$. The curves for $\lambda \lesssim 1$ and $\lambda \gtrsim 3$ for an isotropic viscosity tensor are very similar to the corresponding curves where only $\nu_2$ is varied. In this parameter range the coefficient $\nu_2$ dominates the behavior. Note that the influence of $\nu_3$ on the critical values is already much smaller than that of $\nu_2$. We left out the equivalent graphs for the other viscosity coefficients, because they have almost no effect on the critical values. For further comments on the influence of an anisotropic viscosity tensor see also Sect. 3.4.

All the parameters we have discussed up to now caused variations in the critical values that did not select specific values of the considered parameter. In this aspect the situation is completely different in the case of the flow alignment parameter $\lambda$. As shown in Fig. 9 there is a clear change in behavior for $\lambda \approx 1$ and $\lambda \approx 3$. The
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critical tilt angle is increased for values of $\lambda$ in this interval and the critical wave vector tends to rise only at the boundaries of the interval and is reduced in between. Figure 9 illustrates how this structure depends on the viscosities (assuming all five viscosities to be equal) and on the elastic constants of the layers. In the first row we follow this behavior for viscosities varying from $\nu_i = 1$ down to $\nu_i = 10^{-3}$. Clearly, the influence of $\lambda$ is more pronounced the lower the viscosities are. Both elastic constants of the layers, the compressibility $B_0$ and the bending modulus $K$ (in our dimensionless units $B_1 = 1$), have in general a similar influence on the shape of the graphs: the smaller the elastic constants, the more pronounced the structure becomes. For this reason we just give the plot for $B_0$ (second row in Fig. 9) and omit the plot for $K$.

These dependencies on the system parameters give some important hints for an interpretation of Fig. 9. The currents and quasi-currents for the velocity field and the director consist of two parts (see (17) and (18)): a diagonal one (coupling, e.g., the components of $v$ among each other) and an off-diagonal one (coupling the director to the velocity field). The former is proportional to the elastic constants or to the viscosity tensor whereas the latter is a function of the flow alignment parameter. So reducing either the elastic constants or the viscosities increases the portion of the cross-coupling terms in theses equations, i.e., the observed tendencies are exactly what one would expect. The next step in the interpretation of the shape of the curves is to have a closer look at the structure of the cross-coupling term. The flow alignment tensor $\lambda_{ijk} = \frac{1}{2} \left[ (\lambda - 1) \delta_{ij} n_k + (\lambda + 1) \delta_{ik} n_j \right]$ obviously changes its behavior for $\lambda = 1$: the first part changes its sign. Note that we are in a region of the parameter space where $\lambda_{ijk}$ is a dominating term (since either the viscosities or the elastic constants are small). Additionally, $\delta_{ij} n_k$ contains up to the third power of one director component. For this reason we expect that – in the linearized set of equations – some coupling terms change their sign for $\lambda = 1$ others for $\lambda = 3$. For example, the $\phi$-component of the director is coupled to the $x$ and $z$ components of the velocity field by the terms $(\lambda - 1)/2 \partial_\theta v_x$ and $(\lambda - 1)/2 \cot(\theta_0) \partial_\theta v_z$. Similarly the reversible part of the coupling of $v_y$ to $\phi$ vanishes for $\lambda = 3$. The monitored structure in the plots cannot be attributed to one single cross-coupling term, but the given examples demonstrate that something should happen in this parameter range.

3.2.3 Including the Order Parameters

In the preceding paragraphs we investigated undulations assuming a constant modulus of the order parameter $S_{0(n)} = S_{0(n)}^{(n,s)} + S_{0(n)}^{(n,s)}$. In general one would expect that the undulations in the other observable quantities should couple to some extent to the order parameter. In the formulation of the free energy (see Sect. 2.2) we have assumed that $S_{0(n,s)}$ varies only slightly around $S_{0(n,s)}^{(n,s)}$ and thus only the lowest order terms in $s_{0(n,s)}$ contribute to the free energy. For the spatially homogeneous state we had (see (39) and (41)) a correction to the nematic $S_{0(n)}$ proportional to the square of the shear rate ($\theta_0 \sim \dot{\gamma}$ for low $\dot{\gamma}$):
Evaluating (41) and (45) at onset gives an important restriction on the range of possible parameter values (here the cases of $\alpha^{(n)}$ and $\beta^{(n)}_\parallel - \beta^{(n)}_\perp$). Note that the critical $\theta_0$ is a function of the material parameters

$$s_0^{(n)} = -\frac{2}{\lambda + 1} \frac{B_1}{\gamma_1} \frac{\beta^{(n)}_\parallel - \beta^{(n)}_\perp}{\alpha^{(n)} L_0} \theta_0^2 + O(\theta_0^4)$$

As a consequence $s_0^{(n)}$ must be small compared to $S_0^{(n,s)}$ (which is by construction limited to the range $0 \leq S_0^{(n,s)} \leq 1$). Thus a reasonable restriction is

$$|s_0^{(n)}| \lesssim 0.5$$

As shown in Fig. 10, evaluating this relation at the onset of the instability reduces significantly the physically reasonable range for some parameters. This restriction applies only for the nematic material parameters and, in general, nothing can be said about the corresponding smectic parameters. We will, however, take the smectic parameters in the same range as the nematic ones. If not indicated otherwise we used $L^{(n,s)}_0 = 0.1$, $L^{(n,s)}_\perp = 0.01$, $L^{(n,s)}_\parallel - L^{(n,s)}_\perp = 0.005$, $M_0 = 10^{-4}$, $\beta^{(n,s)}_\perp = 0.01$, $\beta^{(n,s)}_\parallel - \beta^{(n,s)}_\perp = 0.005$, $\alpha^{(n,s)} = 0.001$ for the plots of this section (along with parameter set specified in the previous section).

The ansatz for $s_1^{(n,s)}$ following (35) reads

$$s_1^{(n,s)} = A^{(n,s)}_{s} \exp \left[ \left( i \omega + \frac{1}{\tau} \right) t \right] \sin(q_z z) \cos(q y).$$

The modulations of $S^{(n,s)}$ in the linear analysis are maximum at the boundaries and in phase with the layer displacement $u$. The sign of the amplitude $A^{(n,s)}_{s}$ depends on the coupling to the velocity field (only the anisotropic part $\beta^{(n)}_\parallel - \beta^{(n)}_\perp$ is relevant) and on the coupling to the director undulations (via $M_{ijk}$, only for the nematic
Out of the material parameters connected with the order parameter, only $\beta_{\parallel}^{(n)} - \beta_{\perp}^{(n)}$ has a measurable effect on the critical values. Some more parameters can influence the amplitudes of the order parameter undulation, namely $L_{\perp}^{(n)}$ and $M_0$ (the latter is only present in the case of the nematic order parameter). All amplitudes have been normalized such that $A_\phi = 1$. Note that the smectic $A_\phi^{(s)}$ has been multiplied by $10^6$ in the right column. For a better comparison we used a log–log scale in the lower left plot and changed the sign of $A_\phi^{(s)}$ in this plot.

In general the critical values are not at all or only very slightly influenced by the coupling to the modulus of the order parameter (see Fig. 11). Figure 11 summarizes the parameters with the largest influence on $A_\phi^{(n,s)}$. In almost all investigated cases the modulation of the nematic order is much larger than in the smectic order. Whether the order is reduced or increased in regions where the layers are compressed depends on the phenomenological constants $\beta_{\parallel}^{(n,s)} - \beta_{\perp}^{(n,s)}$ and $M_0$ which have not been measured up to now.
The above results reveal some interesting features. As shown in Table 3, the modulations of the order parameter change sign under inversion of the $z$-axis. Considering the boundary condition (i.e., taking our ansatz) this leads to the fact that the effect on the modulus of the order parameters is maximum at the boundaries. So the linear analysis predicts that the regions where the order parameter is influenced most by the undulations are close to the boundaries. Since the probability for the formation of defects is higher in places where the order parameter is lower, we have identified areas where the creation of defects is facilitated. But our analysis does not allow one to predict the structure of the defects. Nevertheless this effect gives a possible way to reorient the parallel layers. Interestingly, experiments in block copolymers by Laurer et al. [3] show a defect structure close to the boundaries which is consistent with this picture.

### 3.3 Oscillatory Instability

All our arguments in the previous sections were based on the assumption that the undulations set in as a stationary instability. That is, that the oscillation rate $\omega$ in our ansatz (35) vanishes at onset. In this section we will discuss the situation for non-zero $\omega$ and find that our previous assumption was justified. In our linear analysis enters now (for the first time in this chapter) the mass density of the system, which we will choose to be equal to unity $\rho = 1$.

The search for a possible oscillatory instability is slightly different from the procedure used in the stationary case. The solvability condition of the linearized set of equations determines both the neutral curve and the frequency along this curve. When searching for such a solution we scanned approximately the same parameter space as used for Figs. 5–7. Since the frequency tends to zero when the oscillatory neutral curve gets close to the stationary one, we concentrated on the frequency range $0 \leq \omega \leq 2$ and checked in some cases for higher frequencies.

It turned out that only in cases when the director field is very weakly coupled to the layering a neutral curve for an oscillatory instability is possible. This weak coupling manifests itself in small $B_1$ and $\gamma_1$, which is in our set of dimensionless variables equivalent to large $B_0$ and $\nu_i$. Oscillatory neutral curves were only found for $B_0 \gtrsim 100$ or $\nu_i \gtrsim 1$. In all investigated cases an oscillatory neutral curve is either absent or lies above the neutral curve for a stationary instability. When an oscillatory neutral curve is possible, it ends in the points where it meets the stationary neutral curve (see Fig. 12). The corresponding frequency approaches zero in the end points of the oscillatory neutral curve. If we ignore for the moment the stationary neutral curve and consider only the oscillatory instability, the corresponding critical values are found to be rather close to the stationary one and approach each other the weaker the coupling between the director and the layers becomes. To summarize, an oscillatory instability was not found to be possible in all investigated cases and seems to be extremely unlikely to occur.
In most parts of the scanned parameter space no possibility for an oscillatory instability was found. If the director field is only very weakly coupled to the layering (in this plot we used $B_0 = 200$ and $\nu_i = 0.4$) a neutral curve for an oscillatory instability \textit{(dashed line)} appears above the stationary neutral curve \textit{(solid line)}. Note that the critical wave vectors are close to each other for both oscillatory and stationary instability. The \textit{inset} shows the frequency along the neutral curve.

### 3.4 Anisotropic Viscosity

In Fig. 8 we have illustrated that a small viscosity coefficient $\nu_2$ facilitates the onset of undulations. In this section we will have a closer look at the effect of an anisotropic viscosity tensor and ask whether undulations can be caused only due to viscosity effects without any coupling to the director field (i.e., we consider standard smectic A hydrodynamics in this section).

Let us start our considerations by looking at the spatially homogeneous state. In a sample with parallel alignment the apparent viscosity is $\nu_3$, which can easily be seen from the force on the upper boundary:

$$F_\parallel = \hat{e}_z \cdot \sigma = \dot{\gamma} \nu_3 \hat{e}_x \tag{47}$$

Similarly the viscosity of a perpendicular alignment is given by $\nu_2$:

$$F_\perp = \hat{e}_z \cdot \sigma = \dot{\gamma} \nu_2 \hat{e}_x \tag{48}$$

For $\nu_2 < \nu_3$ a simple shear flow in a perpendicular alignment causes less dissipation than in a parallel alignment. The next step is to study the stability of these alignments in the linear regime. Following the standard procedure (as described above) we find a solvability condition of the linearized equations which does not depend on the shear rate $\dot{\gamma}$:

$$0 = \left\{ q^2 + \lambda_p \left[ \nu_3 (q^2 - q_z^2)^2 + 2 (\nu_2 + \nu_3) q^2 q_z^2 \right] \right\} \times \left( B_0 q_z^2 + K q^4 \right) (\nu_2 q_z^2 + \nu_3 q_z^2) \tag{49}$$
Consequently, a parallel alignment of smectic layers is linearly stable against undulations even if the perpendicular alignment might be more preferable due to some thermodynamic considerations. As we have shown in Fig. 8, this rigorous result of standard smectic A hydrodynamics is weakened in our extended formulation of smectic A hydrodynamics. When the director can show independent dynamics, an appropriate anisotropy of the viscosity tensor can indeed reduce the threshold values of an undulation instability.

4 Comparison to Experiments and Simulations

In the previous sections we have shown that the inclusion of the director of the underlying nematic order in the description of a smectic A like system leads to some important new features. In general, the behavior of the director under external fields differs from the behavior of the layer normal. In this chapter we have only discussed the effect of a velocity gradient, but the effects presented here seem to be of a more general nature and can also be applied to other fields. The key results of our theoretical treatment are a tilt of the director, which is proportional to the shear rate, and an undulation instability which sets in above a threshold value of the tilt angle (or equivalently the shear rate).

Both predictions are in agreement with experimental observations. For side-chain liquid crystalline polymers Noirez [25] observed a shear dependence of the layer thickness. In the parallel orientation the layer thickness is reduced by several percent with increasing shear. To our knowledge, two groups have investigated the evolution of a parallel alignment to the vesicle state for lyotropic systems (see Müller et al. [17] and Zipfel et al. [55]). In both papers the authors argue that cylindrical structures (with an axis along the flow direction) are observable as intermediates. These observed cylindrical intermediates are very close to the undulations proposed by our theoretical treatment.

For an approximate quantitative comparison of our theoretical results with the experiments on lyotropic liquid crystals we make a number of assumptions about the material parameters. As we have shown in Sect. 3.2 the different approaches cause only small variations in the critical wave number. For this estimate it suffices to use the critical wave number obtained in our earlier work [42]. For lyotropics it is known [56, 57], that the elastic constants can be expressed as

\[ K = \frac{\kappa}{l} \] (50)

and

\[ B = \frac{9}{64} \pi^2 \left( \frac{k_B T}{\kappa} \right)^2 \frac{l}{(l - \delta)^4}, \] (51)
where $\kappa = \alpha_\kappa k_B T$ is the bending modulus of a single bilayer, $l$ is the repeat distance, $\delta$ is the membrane thickness, $k_B$ is the Boltzmann constant, $T$ is temperature, and $\alpha_\kappa$ is a dimensionless number of order of unity. With this relation we can estimate the critical wave vector for a sample of thickness $d$ using [42]:

$$q_c^2 \approx \frac{3\pi^2}{8\alpha_\kappa d} \frac{l}{(l - \delta)^2}$$

(52)

The parameters of the experiment by Zipfel et al. [55] are: $d = 1$ mm, $\delta = 2.65$ nm, $l = 6.3$ nm, and $\alpha_\kappa = 1.8$ [55,58]. On this basis we estimate the critical wave length to be of the order of

$$\lambda_c \approx 6.4 \, \mu m$$

(53)

Zipfel et al. [55] observed a vesicle radius of 3 $\mu$m, which is clearly compatible with our calculation. We note that this estimate assumes that the experiments are done in the hydrodynamic regime.

In Sect. 3.2.3 we have pointed out that the effect on the order parameter is maximum close to the boundaries of the layer. In a reoriented sample Laurer et al. [3] have identified defects near the boundary of the sample which are in accordance with the predicted influence on the order parameter.

The mechanism we have proposed here is somewhat similar to a shear induced smectic-$C$ like situation. Consequently, undulations should also be observed near the smectic-$A$–smectic-$C$ transitions. Indeed, Johnson and Saupe [59] and later Kumar [60] report such undulations just below the transition temperature. In the same spirit Ribotta and Durand [61] report a compression induced smectic-$C$ like situation.

Molecular dynamic simulations recently made by Soddemann et al. [52,53] offer a very precise insight into the behavior of the layered systems under shear. As we will briefly discuss now, a direct comparison of these simulations to the analytic theory presented above shows very good agreement between both approaches [53]. In Fig. 13 we have plotted the strain rate, $\dot{\gamma}$, as a function of the tilt angle, $\theta_0$.

Clearly flow aligning behavior of the director is present and $\theta_0$ increases linearly with the tilt angle, $\theta_0$. Above a threshold in the strain rate, $\dot{\gamma} \approx 0.011$, undulations in vorticity direction set in. In Fig. 14 the results of simulations for $\dot{\gamma} = 0.015$ are shown. In Fig. 15 we have plotted the undulation amplitude obtained as a function of the shear rate. The dashed line indicates a square root behavior corresponding to a forward bifurcation near the onset of undulations. This is, indeed, what is expected, when a weakly nonlinear analysis based on the underlying macroscopic equations is performed [54]. In Fig. 16 we have plotted an example for the dynamic behavior obtained from molecular dynamics simulations. It shows the time evolution after a step-type start for two shear rates below the onset of undulations. The two solid lines correspond to a fit to the data using the solutions of the averaged linearized form of (27). The shear approaches its stationary value for small tilt angle (implied by the use of the linearized equation) with a characteristic time scale $\tau = \gamma_1/B_1$. 
Fig. 13  Strain rate as a function of tilt angle. This form of presenting the data has been chosen to facilitate a direct comparison with the theory, especially (38). The solid line is a fit to the data. Extracted from Fig. 5.4 of [54]

Fig. 14  Undulations in a simulated model system. At a strain rate of $\dot{\gamma} = 0.015$ clearly undulations have developed. As considered in the theory, undulations in the vorticity direction are present. Note that the undulation amplitude does not change along the $z$-direction. Extracted from Fig. 5.5 of [54]
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**Fig. 15** Undulation amplitude due to shear. The amplitude of the undulations $A$ is given as a function of the strain rate $\dot{\gamma}$. At a shear rate $\dot{\gamma} \approx 0.01$ undulations set in. The amplitude of these undulations grows continuously with increasing shear rate. The *dashed line* shows a fit to data points starting at $\dot{\gamma} > 0.02$ assuming a square root dependence of the amplitude above the undulation onset. Fig. 5.6 of [54]

![Graph showing undulation amplitude vs. strain rate](image1)

**Fig. 16** Time evolution of the director tilt after a step-like start of the shear for two different final shear rates (0.008 and 0.010 in Lennard-Jones units). The lines show the fit to the data using the solution of the averaged linearized form of (27). Fig. 5.12 of [54]

![Graph showing director tilt vs. time](image2)

A detailed analysis of the simulation data [53] demonstrated that the analytic theory is in qualitative and frequently even in quantitative agreement with the simulations over the validity range of the linearized theory.

## 5 Transition to Onions

In this section, we will explore the possibility of having a cylindrical intermediate between the initial lamellar and the final onion state. This possibility has been considered since the shearing experiments of Richtering’s group [55, 62] on lyotropic
lamellar phases exhibiting a shear-induced lamellar-to-vesicle (onion) transition. Performing viscosity measurements accompanied by time-resolved small-angle neutron and light scattering, they observe a formation of an intermediate structure exhibiting a scattering pattern corresponding to objects with a cylindrical symmetry oriented in the flow direction. They suggest two possible structures consistent with the overall cylindrical scattering symmetry: multilamellar cylinders with the long axis oriented along the flow, or large amplitude layer undulations with the wave vector in the neutral (vorticity) direction [62].

In what follows, we will explore the first of the two suggested scenarios. Starting with a multilamellar cylinder configuration, we will study its stability under shear flow (Fig. 17). Our aim is to check whether we can find an instability of the cylinder—a secondary instability—that would be responsible for the break-up into onions.

The strategy is as follows. We start by rewriting the equations in cylindrical coordinates \((r, \phi, z)\). The variables we consider are the layer displacement \(u\) (now in the radial direction) from the cylindrical state, the director \(\hat{n}\), and the fluid velocity \(v\). The central part of the cylinder, \(r < R_1\), containing a line defect, is not included. It is not expected to be relevant for the shear-induced instability. We write down linearized equations for layer displacement, director, and velocity perturbations for a multilamellar (smectic) cylinder oriented in the flow direction \((z\) axis). We are interested in perturbations with the wave vector in the \(z\) direction as this is the relevant direction for the hypothetical break-up of the cylinder into onions. The unperturbed configuration in the presence of shear flow (the ground state) depends on \(r\) and \(\phi\) and is determined numerically. The perturbations, of course, depend on all three coordinates. We take into account translational symmetry of the ground state in the \(z\) direction and use a plane wave ansatz in that direction. Thus, our ansatze for the perturbed variables are
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\[ u = u^{(0)}(r, \phi) + u^{(1)}(r, \phi, z, t), \]  
\[ \hat{n} = \hat{n}^{(0)}(r, \phi) + \hat{n}^{(1)}(r, \phi, z, t), \]  
\[ v = v^{(0)}(r, \phi) + v^{(1)}(r, \phi, z, t), \]

where

\[ u^{(1)} = u_0^{(1)}(r, \phi, t)e^{ikz}, \]  
\[ \hat{n}^{(1)} = \hat{n}_0^{(1)}(r, \phi, t)e^{ikz}, \]  
\[ v^{(1)} = v_0^{(1)}(r, \phi, t)e^{ikz}, \]

and \(u_0^{(1)}, \hat{n}_0^{(1)},\) and \(v_0^{(1)}\) are complex perturbation amplitudes. We use the numerically obtained ground state and evolve the perturbation amplitudes with a selected wave vector in \(z\) direction numerically.

The layer normal \(\hat{p}\), in first order with respect to \(u\), is

\[ \hat{p} = \hat{e}_r - \hat{e}_\phi \frac{\partial u}{\partial \phi} - \hat{e}_z \frac{\partial u}{\partial z}. \]  

Bend of the layers can be conveniently expressed as splay of the layer normal. The bending term (the first term in (3)) is

\[ \frac{1}{2}K (\nabla \cdot \hat{p} - \nabla \cdot \hat{e}_r)^2 = \frac{1}{2}K \left( \frac{\partial^2 u}{r^2 \partial \phi^2} + \frac{\partial^2 u}{\partial z^2} \right)^2, \]

where we have made the expansion around the curved (cylindrical) ground state. Layer compression (the second term in (3)) to the same order is

\[ \frac{1}{2}B_0 \left\{ \frac{\partial u}{\partial r} + n_r \left[ 1 - \frac{1}{2} \left( \left( \frac{\partial u}{r \partial \phi} \right)^2 + \left( \frac{\partial u}{\partial z} \right)^2 \right) \right] \right. \]

\[ + n_\phi \frac{\partial u}{r \partial \phi} + n_z \frac{\partial u}{\partial z} - \frac{1}{2} \left( \left( \frac{\partial u}{r \partial \phi} \right)^2 + \left( \frac{\partial u}{\partial z} \right)^2 \right) \left\} \right. \]

where we have omitted terms containing \(u/r\) or \(\partial u/\partial r\) that are smaller than the first term in (62). Finally, the coupling term (the last term in (3)) is

\[ \frac{1}{2}B_1 \left[ \left( n_z \frac{\partial u}{r \partial \phi} - n_\phi \frac{\partial u}{\partial z} \right)^2 + \left( n_z + n_r \frac{\partial u}{\partial z} \right)^2 + \left( n_r \frac{\partial u}{r \partial \phi} + n_\phi \right)^2 \right] \]

Dynamics of \(u\) and \(\hat{n}\) is then given by (16) and (17). For the boundary condition we take \(u(R_1) = 0\) at the cut-off radius \(R_1\), and \(\frac{\partial u}{\partial r}(R) = 0\) at the surface of the cylinder. It is worth remembering that the functional derivative (14) is rather lengthy.
in cylindrical coordinates. For the form of $\varepsilon$ as given in (61), (62), and (63), the nonzero terms are

$$
\Psi = - \frac{\partial}{\partial r} \frac{\partial \varepsilon}{\partial (\frac{\partial u}{\partial r})} - \frac{1}{r} \frac{\partial}{\partial r} \frac{\partial \varepsilon}{\partial (\frac{\partial u}{\partial \phi})} - \frac{\partial}{\partial z} \frac{\partial \varepsilon}{\partial (\frac{\partial u}{\partial z})}
- \frac{1}{r} \frac{\partial}{\partial r} \frac{\partial \varepsilon}{\partial (\frac{\partial^2 u}{\partial r^2})}
+ \frac{\partial^2}{\partial \phi^2} \frac{\partial \varepsilon}{\partial (\frac{\partial^2 u}{\partial r^2 \partial \phi^2})}
+ \frac{\partial^2}{\partial z^2} \frac{\partial \varepsilon}{\partial (\frac{\partial^2 u}{\partial z^2})}.
$$

(64)

The velocity is calculated from (18) using a series of approximations. We assume that in the ground state the flow is simple shear flow:

$$
v^{(0)} = \dot{\gamma} r \sin \phi \hat{e}_z,
$$

(65)

where $\dot{\gamma}$ is the typical shear rate of the experiment. In our first attempt, we completely neglect the velocity perturbation. The dynamics of layers is thus given solely by permeation (16). This results in a static stability analysis and therefore the fact that permeation is tiny in reality is irrelevant. No instability of the cylinder was found in this case.

From now on, the permeation in (16) is neglected as it is several orders of magnitude smaller than the advection due to the radial component of the velocity $v_r$ (now playing the role of $v_z$ in the planar case). As far as the velocity perturbation is concerned, our aim is to describe its principal effect—the radial motion of smectic layers, i.e., instead of diffusion (permeation) we now have advective transport. In this spirit we make several simplifications to keep the model tractable. The backflow—flow generation due to director reorientation—is neglected, as well as the effect of anisotropic viscosity (third and fourth line of (19)). Thereby (19) is reduced to the Navier-Stokes equation for the velocity perturbation, which upon linearization takes the form

$$
\rho \left( \frac{\partial v_i^{(1)}}{\partial t} + v_z^{(0)} \frac{\partial v_i^{(1)}}{\partial z} \right) = - \nabla_i P^{(1)} - \nabla_j \psi_j^{(1)} \delta_{i1} + \eta \nabla^2 v_i^{(1)},
$$

(66)

where $-\nabla_j \psi_j^{(1)} = \Psi^{(1)}$ is the force density in the radial direction ($i = 1$) and $\eta$ is the isotropic viscosity. The $z$ dependence is already captured by the plane wave ansatz (57)–(59), so $\frac{\partial}{\partial z} = ik$.

Several reduced models for the velocity perturbation $v^{(1)}$ have been systematically tested.

1. Simplest model. The incompressibility condition is not obeyed, the velocity perturbation is stationary (the time derivative in (66) is neglected) and has only a radial direction,

$$
v^{(1)} = v_0^{(1)}(r, \phi, t) e^{ik \hat{e}_z},
$$

(67)

with the solution
Thus, the $r$ and $\phi$ components of the velocity gradient are completely disregarded and $v_0^{(1)}$ depends on those coordinates only through the $r$ and $\phi$ dependence of the (radial) force density and the ground state shear velocity. The quality of this model increases with increasing $kR$. It is expected that the velocity perturbation is overestimated in this model and thus the hypothetical instability threshold is lowered, which makes the model appealing at least as a first attempt.

2. Improved model. The incompressibility is obeyed. The velocity perturbation is stationary and lies in the $r\phi$ planes:

$$v^{(1)} = \left[ v_{0r}^{(1)}(r, \phi, t) \hat{e}_r + v_{0z}^{(1)}(r, \phi, t) \hat{e}_z \right] e^{ikz}$$

(69)

The coordinate $\phi$ is again only a parameter, i.e., all derivatives with respect to $\phi$ are neglected. Hence we are left with the one-dimensional problem

$$\rho v_z^{(0)} i k v_0^{(1)} = -(\nabla_r + ik \hat{e}_z)F_0^{(1)} + \eta (\nabla_r^2 - k^2)v_0^{(1)} + \Psi_0^{(1)} \hat{e}_r$$

(70)

and

$$(\nabla_r + ik \hat{e}_z) \cdot v_0^{(1)} = 0$$

(71)

to solve at every $\phi$ coordinate. Taking the curl of (70) to eliminate the pressure and omitting all $\phi$ derivatives, one finally gets

$$\rho v_z^{(0)} i k \left( ikv_0^{(1)} - \frac{\partial v_{0z}^{(1)}}{\partial r} \right) - \rho \frac{\partial v_z^{(0)}}{\partial r} ikv_0^{(1)} =$$

$$\eta \left\{ \left[ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial}{\partial r} \right) - \left( k^2 + \frac{1}{r^2} \right) \right] \left( ikv_{0r}^{(1)} - \frac{\partial v_{0z}^{(1)}}{\partial r} \right) \right\} + ik \Psi_0^{(1)}. \quad (73)$$

At the outer boundary we assume $\frac{\partial v_0^{(1)}}{\partial r}(R) = 0$, $\frac{\partial v_{0z}^{(1)}}{\partial r}(R) = 0$ and at the cut-off boundary $v_0^{(1)}(R_1) = 0$, $\frac{\partial v_{0r}^{(1)}}{\partial r}(R_1) = 0$ and at the cut-off boundary $v_{0z}^{(1)}(R_1) = 0$, $\frac{\partial v_{0z}^{(1)}}{\partial r}(R_1) = 0$. The resulting band-diagonal system, (71) and (73), is efficiently solved by the conjugate gradient method [63].

3. Time-dependent velocity model. The approximations are identical to the previous model, except that now the velocity field is not stationary, i.e., the time derivative in (66) is nonzero. This introduces another time scale $\tau_v = \rho R^2 / \eta$ to the problem which could be significant for the instability. Now it is convenient to start with (66) containing the pressure. Using (66), the velocity is updated explicitly in time:

$$\frac{v_0^{(1)} - v_0^{(1)}}{\Delta t} = \frac{1}{\rho} \{ \ldots \}, \quad (74)$$
then it is corrected applying a pressure correction $\delta P$ in an auxiliary step [64]:

$$v_{0}^{\text{new}(1)} = v_{0}^{\ast(1)} - \Delta t \nabla \delta P,$$

(75)

such as to satisfy the incompressibility condition (71)

$$\nabla \cdot v_{0}^{\text{new}(1)} = 0 = \nabla \cdot v_{0}^{\ast(1)} - \Delta t \nabla^2 \delta P.$$

(76)

Thus at every time step a Poisson equation for the pressure correction is solved, yielding corrections (75) to the velocity and the pressure, $P_{0}^{\text{new}(1)} = P_{0}^{(1)} + \delta P$.

This leads to the following one-dimensional system:

$$\frac{v_{0r}^{\ast(1)} - v_{0r}^{(1)}}{\Delta t} = \frac{1}{\rho} \left[ -\nabla_r P_{0}^{(1)} + \eta \left( \nabla_r^2 - \frac{1}{r^2} - k^2 \right) v_{0r}^{(1)} + \Psi_{0r}^{(1)} \right],$$

(77)

$$\frac{v_{0z}^{\ast(1)} - v_{0z}^{(1)}}{\Delta t} = \frac{1}{\rho} \left[ -ikP_{0z}^{(1)} + \eta \left( \nabla_r^2 - k^2 \right) v_{0z}^{(1)} \right],$$

(78)

$$\left( \nabla_r^2 - k^2 \right) \delta P = \left( \frac{\partial}{\partial r} + \frac{1}{r} \right) v_{0r}^{\ast(1)} + ikv_{0z}^{\ast(1)},$$

(79)

$$v_{0}^{\text{new}(1)} = v_{0}^{\ast(1)} - \Delta t \nabla \delta P,$$

(80)

$$P_{0}^{\text{new}(1)} = P_{0}^{(1)} + \delta P,$$

(81)

which is solved on a staggered grid [64].

We start with the ground state $u^{(0)}$, $\hat{n}^{(0)}$, defined by the simple shear flow $v^{(0)}$, Fig. 17. The principal effect is, as expected, the appearance of a small tilt of the director from the layer normal (flow alignment), predominantly in $z$ direction (Fig. 18). Note that the configuration of layers is also modified by the shear (Figs. 19 and 20), i.e., the cylindrical symmetry is lost. This is analogous to the shear-flow-induced undulation instability of planar layers (wave vector of undulations in the

![Fig. 18](image-url) Ground state flow alignment: director tilt in the $z$ direction $n_z^{(0)}$ at $r = 0.75R$ as a function of the polar angle $\phi$. The tilt is zero at $\phi = 0$ and $\phi = \pi$, where the director points in the neutral (vorticity) direction.
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**Fig. 19** Second order in $\dot{\gamma} \gamma_1 / B_1$ effects: the ground state layer displacement $u^{(0)}$ and $n_z^{(0)}$ at $r = 0.75R$ as functions of the polar angle $\phi$. Note the quadrupolar deformation of the cylinder.

**Fig. 20** Radial profile of $u^{(0)}$ and $n_z^{(0)}$ close to (but not exactly at) $\phi = \pi/2$. Note the regions of dilatation and compression of the layers caused by the flow alignment.

vorticity direction), except that in the case of the cylinder the translational symmetry in the vorticity direction is already broken by construction (in contrast to the planar layers) and hence there exists no instability threshold but rather a continuously growing deformation of the cylinder as the shear rate is increased. In the lowest order of $\dot{\gamma} \gamma_1 / B_1$ it can be shown that

$$n_z^{(0)} = \frac{1}{2} \gamma \frac{\gamma_1}{B_1} (\lambda + 1) \sin \phi,$$

(82)

$$n_{\phi}^{(0)} = \frac{1}{2} \gamma \frac{\gamma_1}{B_1} (\lambda - 1) n_z^{(0)} \cos \phi,$$

(83)

i.e., $n_{\phi}^{(0)}$ is one order smaller than $n_z^{(0)}$. Additionally it can be shown that $u^{(0)}$ is likewise second order in $\dot{\gamma} \gamma_1 / B_1$. Note that in the lowest order the tilt in the $z$ direction (82) agrees with the tilt in the planar case (40).

We randomly perturb the ground state and evolve the perturbation with a given wave vector $k$ in time numerically, searching for any exponential increase of its amplitude which would be a signature of the instability. An example of such a perturbation is presented in Fig. 21. For all velocity field models, the parameter space
is systematically swept. This includes varying the wave vector of the perturbation $k$, the director tilt (shear rate), the ratio $B_1/B_0$, and the viscosity $\eta$. The result is invariably the same: no instability could be found.

Further, we consider the perturbation $s^{(1)}$ of smectic order $s = s^{(0)} + s^{(1)}$ and check whether it could be important for the instability. We take into account the dependence of the constant $B_1$ on smectic order:

$$B_1 = B_1^{(0)} + B_1' s^{(1)}$$  \hfill (84)

To implement this intuitive physical picture, we take into account the smectic order dependence (84) of $B_1$ in (63), add the energy of smectic order variations (5) to our collection of energy contributions, and regard $s^{(1)}$ as another dynamic variable.
with a short relaxation time. Then we repeat the whole procedure of scanning the parameter space and looking for a growing perturbation by the numerical time evolution. Once more the result is: no instability.

The failure of all our attempts to find an instability suggests one of two possibilities: either the cylinders are stable and as such cannot be candidates for the experimentally observed intermediates with the cylindrical scattering symmetry, or an important ingredient of the physics is missing in our description. Repeated reconsiderations of the experimental circumstances, results, and hints of the experimentalists themselves suggest that the cylinders nevertheless remain the strongest candidate for the puzzling transient state. The quest continues and it now appears that a local (in the sense of the energy landscape) instability of the cylinder triggering the formation of onions is unlikely to exist. Therefore it is reasonable to consider another possibility: perhaps it is not a small amplitude instability, but rather a localized large amplitude perturbation that is responsible for the destruction of the cylinder. In other words, the transition to onions may start with a local rupture of smectic layers via previously nucleated layer defects. A schematic picture of the process is just cutting the multilamellar cylinder and then closing the open layers so created to form the multilamellar vesicles. This process is restricted to the vicinity of the cut and leaves the rest of the cylinder unaffected. The description of such processes is nonperturbative and requires a complete description of both the nematic and the smectic phase transition (a complete description of nematic and smectic defects). Moreover, a three-dimensional numerical calculation is necessary in this case.

6 Concluding Remarks

In this chapter we have shown that a modification of the usual smectic hydrodynamics (layer normal and director are no longer forced to be parallel) will lead to a flow aligning behavior and thus to an effective dilatation of the smectic layers. A linear stability analysis shows that above a critical shear rate the flow alignment is strong enough to cause an undulation instability and thus to destabilize the layered structure. We point out that the linearized analysis presented here does not allow one to predict which structure will be stable at shear rates above the critical shear rate. To overcome this problem, two strategies can be followed. Either one expands the governing equations in small, but non-vanishing amplitudes (in the vicinity of the threshold) along the lines of the work by Schlüter, Lortz and Busse [65] and Newell and Whitehead [66], or one attacks the full non-linear equations by direct numerical integration. The former procedure results in a hierarchy of equations which have to be solved successively leading at a certain order to an envelope equation for the amplitude. This procedure is from the large field of pattern formation and pattern selection in dissipative systems. For recent overviews to this broad field compare, for example, [67–69].
Following the lines proposed above will give a prediction of the pattern formed above onset. For a transition from undulating lamellae to reorientated lamellae or to multilamellar vesicles, defects have to be created for topological reasons. Since the order parameter varies spatially in the vicinity of the defect core, a description of such a process must include the full (tensorial) nematic order parameter as macroscopic dynamic variables.

An interesting similarity of what we discussed here appears if one deals with mixtures of rodlike and disklike micelles. These systems could behave very similarly to a truly biaxial nematic, but show interesting differences to them. Whereas for the usual orthorhombic biaxial nematics both directors are perpendicular to each other by construction, in mixtures there is no need to impose this restriction. Pleiner and Brand [70] investigated how mixtures are influenced by an external field (magnetic field or shear flow) and found that the angle between the two directors exhibits a flow aligning behavior similar to the one studied in \[42, 43\].

Experiments by Müller et al. [17] on the lamellar phase of a lyotropic system (an LMW surfactant) under shear suggest that multilamellar vesicles develop via an intermediate state for which one finds a distribution of director orientations in the plane perpendicular to the flow direction. These results are compatible with an undulation instability of the type proposed here, since undulations lead to such a distribution of director orientations. Furthermore, Noirez [25] found in shear experiment on a smectic A liquid crystalline polymer in a cone-plate geometry that the layer thickness reduces slightly with increasing shear. This result is compatible with the model presented here as well.

In addition, we have investigated here the question of the lamellar to onion (multilamellar vesicles, MLV) transition. This transition was found experimentally [62] to proceed via two possible structures compatible with the overall scattering geometry: either via multilamellar cylinders with the long axis oriented along the flow, or via large amplitude layer undulations with the wave vector parallel to the vorticity direction. Our detailed stability analysis of a multilamellar cylinder shows that such a structure under shear flow is stable against small amplitude perturbations. From our investigations we conclude that the transition from a lamellar structure to onions proceeds via finite amplitude perturbations and/or a local rupturing of the layering. This result is in agreement with very recent experimental results by Koschoreck et al. [20, 21], using small angle neutron scattering, small angle light scattering, and optical microscopy, who showed that lamellar domains can coexist with onions (similar to a two phase region near equilibrium) and that there is a discontinuous growth of vesicle size. All these findings point towards a hysteretic lamellar to onion transition.

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Appendix, Minimal Analytic Model

In our earlier work [42] we considered two independent variables: the layer displacement and the $y$-component of the director. To compare our present analysis to these results we expand (43) and (44) in power series in $\theta_0$ (up to $\theta_0^2$) and take only the terms connected with $\phi$ and $u$:

$$0 = A\phi \frac{B_1}{\gamma_1} \theta_0 - A u \frac{B_1}{\gamma_1} q$$

$$0 = -A u \lambda_p \left[ B_1 q^2 + K q^4 + B_0 q^2 z - \frac{1}{2} \theta_0^2 q^2 (B_0 + 2B_1) \right] + A\phi \lambda_p B_1 \theta_0 q$$

The solvability condition of (85) and (86) defines the neutral curve $\theta_0(q)$ and its minimum directly gives the critical values $\theta_c$ and $q_c$ (within the approximations of this section):

$$q_c^2 = q_z \sqrt{\frac{B_0}{K}}$$

$$\theta_c^2 = 4 q_z \frac{B_0}{B_0 + 2B_1} \sqrt{\frac{K}{B_0}}$$

$$\gamma_c = 4 \frac{B_1}{\gamma_1 (\lambda + 1)} \sqrt{q_z \frac{B_0}{B_0 + 2B_1} \sqrt{\frac{K}{B_0}}}$$

The differences between [42] and (87)–(89) are mainly due to the correct normalization of $\hat{p}$ (see (29)–(31)) used in the present chapter. To summarize, we conclude that our former results are a special case of the present analysis.

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