The Undulation Instability in Layered Systems under Shear Flow - A Simple Model

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ABSTRACT

We propose a generalization to the usual hydrodynamic description of smectic A liquid crystals: The layer normal and the director are assumed to be coupled elastically to each other in such a way that they are parallel in equilibrium. If a sample with the layers parallel to the plates is sheared, our generalization leads in first order to a flow alignment of the director in the flow direction without perturbing the layers. Due to this tilt of the director the layers have a tendency to reduce their thickness. A linear stability analysis shows that the layers accommodate this tendency above a critical shear rate by developing undulations with a wave vector parallel to the vorticity direction of the shear flow.

KEYWORDS

shear flow, smectic A liquid crystals, undulation instability, layered systems

I. INTRODUCTION

Over the last decade a large number of experiments has been performed on the reorientation behavior of layered systems under shear flow. The systems under investigation include low molecular weight liquid crystals exhibiting a smectic A phase (Panizza et al. 1995, Safinya et al. 1991), lyotropic lamellar phases (Diat et al. 1993, Diat and Roux 1993), liquid crystalline side chain polymers (Noirez et al. 1997a, Noirez and Lapp 1997b) and block copolymers (Wiesner 1997, Zipfel et al. 1999). These experiments revealed a variety of interesting flow characteristics which are each common to several types of these layered systems, including reorientation of the layers and the formation of onion-like structures. Typically, at low shear rates the layers are oriented parallel to the plates ("parallel" orientation). At higher shear rates this orientation becomes unstable and multilamellar vesicles ("onions") or layers normal to the vorticity direction ("perpendicular" orientation) are observed. For even higher shear rates some systems show another regime where the stable orientation of the layers is once again parallel to the plates (Diat et al. 1993, Wiesner 1997, Leist et al. 1999). These systems exhibit, however, some interesting differences. Onions are usually observed in systems in which the layer thickness is increased by a solvent, as in lyotropic lamellar phases (either made of surfactants — see the work of Roux, Diat and coworkers — or of block copolymers — see Zipfel et al. 1999). In block copolymer melts (Koppi et al. 1992, Winey et al. 1993a and 1993b, Zhang et al. 1995, Wiesner 1997, Leist et al. 1999) experiments are usually carried out under large amplitude oscillatory shear. Recently Leist et al. 1999 have shown, that the reorientation can be described as a function of the shear rate rather than as function of the oscillation frequency. In thermotropic low molecular weight liquid crystals and liquid crystalline polymers only the first two regimes are observed (Panizza et al. 1995, Noirez et al. 1997a).

Using the usual set of hydrodynamic equations of the smectic A phase (Martin et al. 1972, de Gennes and Prost 1993, Pleiner and Brand 1996) the observed change of orientation cannot be explained, because in these models each layer is assumed to be a two dimensional fluid. In the framework of irreversible thermodynamics (de Groot and Mazur 1969, Forster 1975) the macroscopic variables of a system can be divided in those due to conservation laws (here: mass density ρ, momentum density $\vec{g} = \rho\vec{v}$ with the velocity field $\vec{v}$ and energy density ϵ) and those reflecting a spontaneously broken continuous symmetry (here the layer displacement u characterizing 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 a finite angle between $\hat{n}$ and $\hat{p}$ has been shown to be of physical interest (Litster et al. 1979).

Smectic A liquid crystals are known to be very sensitive against dilatations of the layers. As Clark and Meyer (1973) and Delaye et al. (1973) have shown, a relative dilatation of less than 10^{-4} parallel to the layer normal suffices to cause an undulation instability of the smectic layers. Later on Oswald and Ben-Abraham (1982) investigated the influence of an additional steady shear flow on the undulation instability. They have shown that undulations with a wave vector parallel to the vorticity direction sets in at the same threshold value of dilatation as in the static case. For wave vectors with non-vanishing projections on the flow direction the threshold is increased relative to the static case. Thus the instability a smectic A liquid crystal under shear flow exhibits at lowest dilatation is an undulation instability with a single wave vector parallel to the vorticity direction. Over the last decade several explanations have been proposed for specific systems. In 1992 Bruinsma and Rabin considered the effect of shear flow on layer fluctuations in lamellar phases. They 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. Williams and MacKintosh (1994) 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 Clark and Meyer (1973) and Delaye et al. (1973). Very recently Zilman and Granek (1999) considered short wavelength fluctuations. In their model these fluctuations are suppressed for energetic reasons leading to an undulation instability of the layers.

In this contribution 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. Differently from previous approaches we derive the set of macroscopic dynamic equations within the framework of irreversible thermodynamics and perform a linear stability analysis of these equations. The key point in our 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. The paper is organized as follows. In Sect. II we develop the set macroscopic equation describing our model. Sect. III is divided in two parts. In Sect. III A we solve the macroscopic dynamic equations neglecting cross-coupling terms between the layer displacement u and the director $\hat{n}$. This simplified analysis already contains most of the dominant physical results of our model. In Sect. III B we present a more complete linear stability analysis of the governing equations. We demonstrate that an undulation instability is possible assuming that the elastic constants of the system fulfill a certain condition. Finally we discuss the numerical values entering in our model as well as a comparison of our results with recent experiments.

II. THE MACROSCOPIC EQUATIONS

We consider an infinite layer of a mono-domain smectic A liquid crystal of thickness d as shown in Fig. 1. The plates are parallel to the xy-plane, the unperturbed flow is parallel to the x-axis. In the following we call the y-direction the vorticity direction and the plane spanned by the flow direction and the vorticity direction the shear plane. We note, that there are other conventions about the notion "shear plane", where "shear plane" refers to the plane containing the flow direction and the normal of the sample plane. Both plates move with a velocity of $\frac{v_0}{2}$ along the x-axis but in opposite direction, thus giving rise to an average shear rate of $\dot{\gamma} = \frac{v_0}{d}$. Since our considerations are based on the symmetry of the system we use the expressions "layered" and "smectic" as analogous (thus neglecting e.g. internal degrees in polymeric systems). To simplify our notation we will use the following convention. Vectors are either written with an arrow above the symbol or indexed with a letter representing its components (e.g. $\vec{v}$ or v_i for the velocity field, where i stands for x, y or z). If the vector is normalized to unit length it is written with a hat over the symbol (e.g. $\hat{n}$ for the director). In the same way a tensor is given as an underlined symbol or with two lower indices (e.g. $\underline{\sigma}$ or σ_{ij} for the stress tensor). The same convention applies for nabla $\nabla = (\frac{\partial}{\partial x}, \frac{\partial}{\partial y}, \frac{\partial}{\partial z})$, where ∇_i stands for one of its components. Summation over repeated indices is always assumed.

Several terms contributing to the energy density reflect the symmetry of the system. The director $\hat{n}$ does not distinguish between head and tail, thus it can only occur quadratically in this energy density. Furthermore the energy density of the system is invariant under rigid rotations. Therefore the lowest order term of the director which can enter in the energy density is of the form $K_{ijkl}(\nabla_j n_i)\,(\nabla_l n_k)$. In this paper we will use the standard formulation

$$\frac{1}{2}K_1(\nabla\cdot\hat{n})^2 + \frac{1}{2}K_2[\hat{n}\cdot(\nabla\times\hat{n})]^2 + \frac{1}{2}K_3[\hat{n}\times(\nabla\times\hat{n})]^2 \tag{1}$$

which represent splay, twist and bend deformations respectively (de Gennes and Prost 1993). Also variations of u only contribute as gradient terms, since rigid translations of the whole system cannot change its energy. In linear approximation, transverse gradients of u correspond to a rotation of the layers (which again cost no energy); therefore one must take into account second order derivatives in the transverse directions. Invariance under parity requires

that u appears quadratically in the energy density. Thus the lowest order terms due to the layered structure can be written as

$$\frac{1}{2}K\left(\frac{\partial^2 u}{\partial x^2}+\frac{\partial^2 u}{\partial y^2}\right)^2+\frac{1}{2}B_0\left(\frac{\partial u}{\partial z}\right)^2, \tag{2}$$

describing the curvature of the layers and their dilatations respectively. As mentioned above, neither rigid rotations of the director field nor rigid rotations of the layers can contribute to the energy density due to rotational invariance, but relative rotations of $\hat{n}$ versus $\hat{p}$ may contribute to the energy. Assuming a small angle between $\hat{n}$ and $\hat{p}$ we write this term as:

$$\frac{1}{2}B_1(\hat{n}\times\hat{p})^2. \tag{3}$$

We note that this term is non-hydrodynamic, since it does not vanish in the limit of small wave number excitations (i.e. $q\to 0$). It thus leads dynamically to a relaxation and not to diffusive behavior in the long wavelength limit. In the following considerations we make several simplifications: 1) Since bend deformations are rather higher order gradient corrections to dilatations, if the angle between $\hat{n}$ and $\hat{p}$ is small, we will neglect bend. 2) In the hydrodynamics of smectics twist deformations are forbidden. Thus, for $\hat{n}$ close to $\hat{p}$, any twist of $\hat{n}$ has to be very small and we will neglect it.

To derive the set of macroscopic equations describing our model we follow the standard procedure (Martin et al. 1972, Forster 1975, Pleiner and Brand 1996). In addition to the energy density discussed above, other key ingredients in this procedure are the Gibbs relation, balance equations for the macroscopic variables (see introduction) and the dissipation function R. Here we will discuss only the necessary extensions to the hydrodynamics of nematic liquid crystals given previously (see Pleiner and Brand 1996 and references therein).

In the Gibbs relation we must take into account, in the spirit of our model, both, the terms due to the nematic order and those due to the smectic order. These terms can be written as

$$\mathrm{d}\epsilon = \mathrm{d}\epsilon_0+\Psi_i\,\mathrm{d}(\nabla_i u)+h'_i\,\mathrm{d}n_i+\Phi_{ij}\,\mathrm{d}(\nabla_j n_i), \tag{4}$$

where $\vec{\Psi}$, $\vec{h}'$ and $\underline{\Phi}$ are the conjugate quantities to ∇u, $\hat{n}$ and $\nabla\hat{n}$, respectively. They are given by the derivatives of the energy density with respect to the appropriate variables (e.g. $\Psi_i=\frac{\partial\epsilon}{\partial(\nabla_i u)}$).

For variables which arise from conservation laws, these laws lead directly to the balance equations:

$$\frac{\partial}{\partial t}\rho+\nabla_i(v_i\rho)=0 \tag{5}$$

$$\frac{\partial}{\partial t}g_i+\nabla_j(v_i g_j)+\nabla_j\sigma_{ij}=0 \tag{6}$$

$$\frac{\partial}{\partial t}\epsilon+\nabla_i[v_i(\epsilon+p)]+\nabla_i j^\epsilon_i=0 \tag{7}$$

$\underline{\sigma}$ denotes the stress tensor, $\vec{j}^\epsilon$ the energy current and p the pressure. Similar balance equations are obtained for the other variables (associated with broken symmetries and the entropy density).

$$\frac{\partial}{\partial t}\sigma+\nabla_i(v_i\sigma)+\nabla_i j^\sigma_i=\frac{R}{T} \tag{8}$$

$$\frac{\partial}{\partial t}n_i+v_j\nabla_j n_i+Y_i=0 \tag{9}$$

$$\frac{\partial}{\partial t}u+v_j\nabla_j u+Z=0 \tag{10}$$

In Eqs. (8-10) σ is the entropy density, T is the temperature and $\vec{j}^\sigma$, $\vec{Y}$ and Z are the (quasi-)currents associated with σ, $\hat{n}$ and u respectively. The pressure is connected with the other macroscopic variables via the Gibbs-Duhem relation

$$p=-\epsilon+\mu\rho+T\sigma+\vec{v}\cdot\vec{g}. \tag{11}$$

Here μ, the chemical potential, is the conjugate quantity to the mass density ρ. The dissipation function can be written as a positive definite quadratic form of the forces:

$$R=\frac{1}{2}\gamma_1^{-1}h_i\delta^\perp_{ik}h_k+\frac{1}{2}\nu_{ijkl}(\nabla_j v_i)\,(\nabla_l v_k)+\frac{1}{2}\kappa_{ij}(\nabla_i T)\,(\nabla_j T)+\frac{1}{2}\lambda_p(\nabla_i\Psi_i)^2, \tag{12}$$

where γ_1 is the rotational viscosity, ν_{ijkl} the viscous stress tensor (consisting of five independent coefficients for a uniaxial system), κ_{ij} the thermal conductivity, λ_p the permeation coefficient, $\delta^\perp_{ij}=\delta_{ij}-n_i n_j$ is the transverse Kronecker symbol and $h_i=\frac{\delta\epsilon}{\delta n_i}=h'_i-\nabla_j\Phi_{ij}$ is the variation of the energy density with respect to the director.

Since in our model $\hat{n}$ and $\hat{p}$ are, in general, not parallel, the thickness of the layers under the influence of an external force need *not* be constant. Whenever $\hat{n}$ and $\hat{p}$ enclose a non-vanishing angle, the projection of $\hat{n}$ on $\hat{p}$ is smaller than unity, thus there is a tendency of the layers to reduce their thickness.

In all balance equations we split the currents and quasi-currents in two parts: a reversible and an irreversible one. Inserting the balance equations into the Gibbs relation allows to determine both, the reversible and the irreversible currents and quasi-currents and leads to a set of macroscopic equations containing several phenomenological coefficients. Symmetry considerations including rotational and translational invariance reduce the number of phenomenological constants further. We find, for example:

$$\sigma_{ij} = p\delta_{ij} + \Phi_{kj}\nabla_i n_k + \Psi_j(\nabla_i u + \delta_{iz}) - \frac{1}{2}\lambda_{kji}h_k - \nu_{ijkl}\nabla_l v_k \tag{13}$$

$$Y_i = -\frac{1}{2}\lambda_{ijk}\nabla_j v_k + \frac{1}{\gamma_1}\delta^{\perp}_{ik}h_k \tag{14}$$

with the flow alignment parameter λ given by

$$\lambda_{ijk} = (\lambda - 1)\delta^{\perp}_{ij}n_k + (\lambda + 1)\delta^{\perp}_{ik}n_j. \tag{15}$$

Note that the transversal Kronecker symbols ensure the normalization of $\hat{n}$.

The macroscopic description of our model contains elements of both, nematic and smectic A hydrodynamics. Their usual descriptions are included as limiting cases in our model, provided we suppress the approximations made in the energy density mentioned above. Letting $B_1 \to \infty$, $\hat{n}$ and $\hat{p}$ are parallel and the equations become those usually used for smectic A. On the other hand, if B_0 and B_1 are zero, one obtains the hydrodynamics of nematic liquid crystals. This does not imply, that our model describes the nematic–smectic A phase transition. To give such a model one has to take into account the nematic and smectic order parameter as additional dynamic macroscopic variables.

III. FLOW ALIGNMENT AND ITS CONSEQUENCES

A. Analysis in Two Separate Steps

We analyze the set of equations in two steps: First we determine the flow field and the director assuming that the layers are unchanged by the shear flow (i.e. they stay parallel to the plates and keep their thickness). In a second step we investigate undulations of $\hat{n}$ and $\hat{p}$ with a wave vector parallel to the vorticity direction.

As described above, the upper plate moves with a velocity $\vec{v}_u = \frac{v_0}{2}\hat{e}_x$ and the lower one with $\vec{v}_l = -\frac{v_0}{2}\hat{e}_x$, thus there is an average velocity gradient of $\dot{\gamma} = \frac{v_0}{d}$ applied to the sample. For the velocity field we assume no-slip boundary conditions at the plates. Throughout our analysis the density ρ and the temperature T are taken to be constant and permeation is neglected. We look for a simple stationary solution of the balance equations. We assume weak anchoring at the boundaries in the sense that the director is free to rotate around its equilibrium orientation (perpendicular to the plates) *without* any energy barrier. This implies that the boundaries have no orienting effect on the director field.

Under the assumption that $\hat{n}$ and u are constant (i.e. $\vec{\Psi} = 0$ and $\Phi_{ij} = 0$) the linear velocity profile

$$\vec{v} = \dot{\gamma} z \hat{e}_x. \tag{16}$$

satisfies linear momentum conservation (6, 13) (de Gennes and Prost 1993). Now we determine the director orientation so that the quasi-current $\vec{Y}$ vanishes. Inserting the above velocity profile in $\vec{Y} = 0$ and supposing an unchanged layered structure leads to the equation

$$\left[\frac{\lambda + 1}{2} - \lambda n_x^2\right]\dot{\gamma} = \frac{B_1}{\gamma_1}n_x n_z + \frac{B_0}{\gamma_1}n_x(1 - n_z), \tag{17}$$

with $n_z = \sqrt{1 - n_x^2}$ and $n_y = 0$. Assuming the angle between $\hat{n}$ and $\hat{p}$ to be small and taking into account only linear terms in n_x, we find[1]

[1]Note that this stationary solution also occurs for $|\lambda| < 1$. The tumbling solution found for nematics for $|\lambda| < 1$ above the nematic-smectic A transition cannot occur in smectic A due to the layering.

$$n_x = \dot{\gamma}\frac{\gamma_1}{B_1}\ \frac{1+\lambda}{2}. \tag{18}$$

As shown in Fig. 2a this result has important consequences: The non-vanishing projection of $\hat{n}$ on the flow direction directly leads to a z-component of the director less than unity.

$$n_z = 1 - \frac{1}{2}n_x^2 + \text{higher order terms} \tag{19}$$

Following the discussion in Sect. II, this tilt of $\hat{n}$ is equivalent to an effective *dilatation* of the layers. Because of this tilt the layers have a tendency to reduce their thickness, but cannot do so due to the boundary conditions.

To analyze the effect of this dilatation we follow the lines of Clark and Meyer (1973) and Delaye et al. (1973) and investigate whether an undulation of the layers can reduce the energy density. In accordance with the results of Oswald and Ben-Abraham (1982) we suppose the wave vector of the undulation to point in the vorticity direction (Fig. 2b).

$$\vec{q} = q_y \hat{e}_y \tag{20}$$

Since we are interested in the onset of undulations of the smectic layers we use the replacement (see Clark and Meyer (1973) and Delaye et al. (1973)).

$$\frac{\partial u}{\partial z} \to \frac{\partial u}{\partial z} - \frac{1}{2}\left(\frac{\partial u}{\partial y}\right)^2 + \text{higher order terms} \tag{21}$$

For small amplitudes undulation of the layers, the angle between the untilted ($\hat{p}_0$) and the tilted layer normal ($\hat{p}$) is given by $\varphi \approx |\hat{p}_0 - \hat{p}| \approx \frac{\partial u}{\partial y}$ (see Fig. 3a). Measured along the z-axis the distance increases by δl compared to the layer thickness l (Fig. 3b). For small φ one can express this apparent dilatation by $\frac{\delta l}{l} \approx \frac{1}{2}\left(\frac{\partial u}{\partial y}\right)^2$. This non-linear correction has turned out to be crucial to explain the undulation instability in dilated smectic A liquid crystals. The undulation amplitude must vanish at the plates, so our ansatz for the layer displacement is (see also Fig. 2b)

$$u = A\cos(\frac{\pi}{d}z)\cos(q_y y) + \frac{1}{2}n_x^2 z, \tag{22}$$

where A is the small amplitude of the undulations, leading to a layer normal of the form

$$\hat{p} = q_y A\cos(\frac{\pi}{d}z)\sin(q_y y)\ \hat{e}_y + \hat{e}_z + \text{higher order terms.} \tag{23}$$

In addition we assume that $\hat{n}$ follows the undulations of $\hat{p}$ in such a way that we can neglect the B_1 term in the energy density for the undulations.

For low shear rates it costs energy to generate undulations, but at higher shear rates the effective dilatations may cause an instability. In between there is a marginal stable state. To determine the parameters of this marginal stable state, we insert the equations (22) and (23) in the simplified version of the energy density discussed in Sect. II. An analysis along the lines of that for dilated smectic A shows that the onset of the instability is given by

$$\frac{1}{2}n_x^2 = \left(\frac{\pi}{d}\right)^2\frac{1}{q_y^2} + \frac{K}{B_0}q_y^2, \tag{24}$$

Minimization with respect to q_y leads to the threshold values:

$$n_{x,c}^2 = 4\ \frac{\pi}{d}\sqrt{\frac{K}{B_0}} \tag{25}$$

$$q_{y,c}^2 = \frac{\pi}{d}\sqrt{\frac{B_0}{K}} \tag{26}$$

$$\dot{\gamma}_c = \frac{4}{1+\lambda}\ \frac{B_1}{\gamma_1}\sqrt{\frac{\pi}{d}\sqrt{\frac{K}{B_0}}} \tag{27}$$

Formulas (25) and (26) are identical to the results obtained in the case a of dilated smectic A phase if one exchanges $\frac{1}{2}n_{x,c}^2$ with the critical dilatation α_c.

B. Analysis of the Combined Set of Equations

In the following we carry out a linear stability analysis of the combined dynamic equations for $\hat{n}$ and u assuming the undulations do not influence the flow field. Taking for u and $\hat{p}$ the ansätze (22) and (23) and allowing $\hat{n}$ to undulate similarly to $\hat{p}$

$$\hat{n} = n_x \hat{e}_x + q_y \tilde{A} \cos(\frac{\pi}{d} z) \sin(q_y y) \hat{e}_y + (1 - \frac{1}{2} n_x^2) \hat{e}_z + \text{higher order terms} \tag{28}$$

we find just minor corrections to the results presented in the previous section. Looking for stationary solutions the equations to be solved are the following.

$$\vec{Y} = 0 \tag{29}$$

$$\nabla \cdot \vec{\Psi} = 0 \tag{30}$$

The x and z components of (29) lead to the same result as equation (18). From the y component of (29) we find that the ratio of the undulation amplitudes contained in $\hat{n}$ and $\hat{p}$ is close to unity.

$$\tilde{A} = \frac{B_1}{B_1 + K q_y^2} \; n_z \; A \tag{31}$$

Inserting typical values (see e.g. Clark and Meyer 1973 and Delaye et al. 1973 for K and q_y and the discussion below for B_1) shows that the fraction in (31) is in a good approximation equal to unity. Inserting this result in (30) we find that the critical wavelength stays unchanged, whereas the critical flow alignment angle acquires a small correction (and so does the critical shear rate)

$$n_{x,c}^2 = 4 \frac{B_0}{B_0 - 2B_1} \; \frac{\pi}{d} \sqrt{\frac{K}{B_0}} \tag{32}$$

$$\dot{\gamma}_c = \frac{4}{1+\lambda} \; \frac{B_1}{\gamma_1} \sqrt{\frac{B_0}{B_0 - 2B_1}} \; \frac{\pi}{d} \sqrt{\frac{K}{B_0}} \tag{33}$$

Before discussing numerical values, we want to point out some important implications of our model (equations (27) and (33)). The critical shear rate increases with increasing B_1. The refinement of our first analysis shows, that no undulation instability is possible if $2B_1$ exceeds B_0. These results could explain why some layered systems do not show a destabilization of the layers parallel to the plates under shear flow (e.g. most thermotropic smectic A liquid crystals far from the phase transition).

For smectic A liquid crystal it is known (Clark and Meyer 1973, Delaye et al. 1973) that α_c is of the order of 10^{-5}, so we expect $n_{x,c}$ to be of the order of 10^{-2}. Thus, there would be only a comparatively small change to the uniaxial nature of a layered system even just below the onset of the undulation instability. To give a numerical value for the critical shear rate appears rather difficult, because neither the elastic constant B_1 nor the rotational viscosity γ_1 are used for the hydrodynamic description of the smectic A phase. Therefore, the only possibility appears to find measurements in the vicinity of the nematic-smectic A phase transition. Measurements on low molecular weight liquid crystals made by Litster et al. (1979) 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 γ_1 we could not find any measurements which would allow an estimate of its value in the smectic A phase. In the nematic phase γ_1 increases drastically towards the nematic-smectic A transition (see e.g. Graf et al. (1992)).

IV. CONCLUDING REMARKS

In this paper 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 (see equation (18) and the discussion thereafter). 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 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 (1963) and Newell and Whitehead (1969). 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 also allows to deal with more than one wave vector for the undulations and thus gives a tool to determine the structure just above onset. This procedure is from the large field of pattern formation and pattern selection in dissipative systems. For recent overviews to this broad field see e.g. Cross and Hohenberg (1993), Buka and Kramer (Eds., 1996) or Busse and Müller (Eds., 1998).

Following the lines proposed above will allow to 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. Both types of refinements (non-linear analysis and inclusion of defects) are beyond the scope of the present paper.

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 (1985) 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 equation (18).

Very recent experiments by Müller et al. (1999) on the lamellar phase of a lyotropic system (a low molecular weight 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 (1999) 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. Nevertheless further investigations on these points are highly desirable.

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FIGURE CAPTIONS

Figure 1: We consider an idealized geometry of a shear experiment. Between two parallel plates we assume a defect-free well aligned lamellar phase. The upper plate moves with the velocity $\frac{v_0}{2}$ in positive x direction, the lower plate moves with the same velocity in negative x direction. The y-direction points into the xz-plane. We call the plane of the plates (xy-plane) the shear plane, the x-direction the flow direction and the y-direction the vorticity direction.

Figure 2: a) In our model the layer thickness is coupled to the director along the layer normal $\hat{p}$. A small but finite angle between $\hat{n}$ and $\hat{p}$ of the order of n_x (due to the flow alignment of the director) reduces the projection of the director onto the layer normal by $\frac{1}{2}n_x^2 l$. b) Schematic picture of the undulations with a strongly exaggerated undulation amplitude. Note the difference in the directions: $\hat{n}$ is tilted in the flow direction, whereas the wave vector of the undulations points in the vorticity direction.

Figure 3: For undulations the layer normal is not parallel to the z-axis, but is tilted by a small angle $\varphi \approx |\hat{p}_0 - \hat{p}| \approx \frac{\partial u}{\partial y}$ in the y-direction. Measured along the z-axis the distance increases by δl compared compared to the layer thickness l. For small φ, δl is given to lowest order by $\frac{\delta l}{l} = \frac{1}{2}\varphi^2$.

Figure 1:

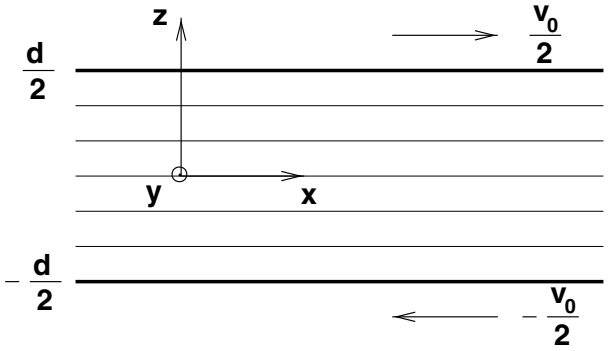

Figure 2:

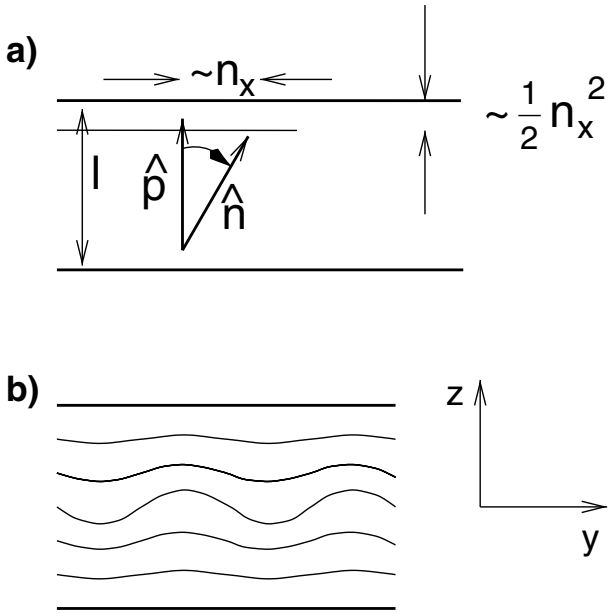

Figure 3: