Instabilities in smectic films*

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Abstract. We discuss various kinds of instabilities in smectic liquid crystal films. Instabilities of the layer structure, within the layer structure, and combined ones can be discriminated.

1 Introduction

Smectic liquid crystals are characterized by a layered structure. In the simplest case the layers are defined by a sinusoidal modulation of the density with a relative amplitude of a few percent [de Gennes and Prost 1993]. This layering constitutes a spontaneously broken translational symmetry in one dimension, quite analogously to crystals with their translational order in three dimensions (and discotics, which have a two-dimensional translational order). Although the positional order of the layers in smectics is not really long-ranged, because of the Landau-Peierls instability [note 1], for practical purposes (reasonable experimental samples) it can be treated as truly long-ranged.

Due to the liquid crystalline nature smectics can be driven out of equilibrium not only by temperature or pressure gradients, or by imposed external flow, also external electric and magnetic fields can be used to obtain instabilities. This allows for completely new instability mechanisms as well as for complex instability scenarios even for the first instabilities. The advantage of films lies in their superior visualization properties and in the possibility to explore new geometries in the experimental setup.

Layering is the basic feature of smectics. In smectics A liquid crystals this is the only order present. In more complex smectics additional (nematic-like) rotational order (smectic C) or bond-orientational order (hexatic B) or both (smectic I and F) exists and in helical smectics (C^*, I^* and F^*) an additional translational order along (but generally incommensurate) to the layers occurs. Of course, there are even more ordered smectic systems, which, however do not yet play a role in pattern formation, since their equilibrium behaviour still has to be investigated.

Instabilities in smectics can be taken in three groups: those, where the layers themselves are subject to distortions and pattern formation, those, where the instability takes place completely in-plane (the layers staying flat), and those, where both kinds of deformations occur simultaneously. We will only briefly comment on the rather classical first case, give a more detailed description of some examples of the second kind and make a few remarks at the end on the still to be explored third case.

2 Layer Instabilities

Layer instabilities in smectics A are well-known and described in the literature [de Gennes and Prost 1993]. There is the undulation instability due an applied mechanical dilation, closely related to the buckling instability in metal plates [Boucif et al 1984] or in discotic systems [Cagnon et al 1984]. Here the system reacts to external constraints by layer undulations rather than by layer dilation, because of energetic reasons. Quite similar undulation instabilities can occur, if a temperature gradient or an electric field is applied along the layer normal (thermo-buckling [Pleiner and Brand 1985] and electro-buckling [Pleiner and Brand 1987]), since in both cases layer dilations can occur [note 2]. Undulation instabilities also arise, if a magnetic or electric field is applied perpendicular to the layers and the magnetic or electric susceptibility anisotropy is positive. Then the layer normals tend to rotate inducing buckling of the layers (Helfrich-Hurault effect [Helfrich and Hurault 1970]). Undulation instabilities in more complex smectics are far less investigated and imply in-plane distortions and pattern forming processes as well.

3 In-Plane Instabilities

Keeping the layers fixed a smectic A liquid crystal behaves like a two-dimensional isotropic liquid within the layers and all the instabilities known from simple liquids can be obtained. An interesting instability type is the so-called vortex-flow instability under a perpendicular electric field due to surface charges or charge separation (although the precise mechanism is still unclear) [Morris et al 1991]. This instability seems to exist in the smectic C phase as well [Becker et al].

A smectic C liquid crystal corresponds to a two-dimensional nematic, if the layers are fixed. Here the standard nematic instabilities, Frederiks transition and electroconvection, can be expected. In addition, rotating mechanical [Cladis et al 1985] and electrical fields [Cladis et al 1995] give rise to interesting target and spiral wave patterns. In the chiralized version, smectic C^*, an in-plane polarisation exists rendering the in-plane system ferroelectric-like. If the twist (the polarization helix) is suppressed or negligible (very thin film), the polarisation exists globally and allows for new coupling effects to an external electric field. This leads to new features in the Frederiks and in the electroconvection instability described below and to target waves in a rotating electric field [Kremer et al 1990], [Hauck and Koswig 1991].

3.1 Polarization Frederiks Transition

We derive first the fully nonlinear equations to investigate director deformations above a DC-driven splay Frederiks-transition in smectics C* within the layers. We are looking for solutions, which are homogeneous perpendicular to the external field. We assume incompressibility and do not take into account the thermal degree of freedom. The backflow is very weak in this situation [Pieranski et al 1973] and will be neglected. With notation and scalings of [Zimmermann et al 1996] we write down the energy density f_G containing a dielectric part f_E and an elastic part f_F due to distortions of the in-plane director $\hat{c}$:

$$f_G = f_F + f_e \tag{1}$$

$$f_e = -\frac{1}{2}\,\epsilon_{ij} E_i E_j - P_i E_i \tag{2}$$

$$f_F = \frac{1}{2} F_{22}\,(\mathrm{div}\,\boldsymbol{c})^2 + \frac{1}{2} F_{33}\,(\boldsymbol{c}\cdot\mathrm{curl}\,\boldsymbol{c})^2 + \frac{1}{2} F_{11} (\boldsymbol{c}\times\mathrm{curl}\,\boldsymbol{c})^2\,, \tag{3}$$

Where the anisotropic tensors are of the form $\epsilon_{ij} = \epsilon_\perp \delta_{ij} + \epsilon_a\, c_i c_j$ and $\epsilon_\perp$ is the dielectric constant perpendicular to the director $\boldsymbol{c}$. F_{22} is the splay, F_{33} the twist and F_{11} the bend elastic constant introduced first in [Saupe 1969]. The molecular field h_i and the dielectric displacement D_i are given by

$$h_i = -\frac{\delta f_G}{\delta c_i} \qquad D_i = -\frac{\delta f_G}{\delta E_i} \tag{4}$$

where the electric field E_i is due to the applied voltage V as well as due to the induced potential. In the given geometry only two degrees of freedom are left, one angle $\theta\,(z,t)$ of the director orientation and the induced electric potential $\phi\,(z,t)$. The nonlinear balance equation of the director is given by

$$\partial_t c_i = \frac{1}{\gamma_1}\,\delta^{tr}_{ik}\, h_k \tag{5}$$

where $\delta^{tr}_{ik} = \delta_{ik} - c_i c_k$. With $c_z = \sin\theta$ this takes the (dimensionless) form:

$$\begin{aligned} \partial_t\theta &= \left((1-c_3^2)\,h_z - c_x c_z h_x\right)/\cos\theta \\ &= \left((\frac{F_{11}}{F_{22}} - 1)(\partial_z\theta)^2 + \frac{\epsilon_a}{\epsilon_\perp}\left(V(t) - \partial_z\phi\,(z,t)\right)^2 \right) \sin\theta\cos\theta \\ &\quad + \left(\frac{F_{11}}{F_{22}}\sin^2\theta + \cos^2\theta \right) \partial_z^2\theta - \sin\theta\; p_0\,\left(V(t) - \partial_z\phi\,(z,t)\right) \end{aligned} \tag{6}$$

The equation of motion for ϕ is derived from the Maxwell equation div$\boldsymbol{D} = \rho$. Eliminating ρ via the charge conservation law $\partial_t\,\rho + \boldsymbol{\nabla}\cdot\boldsymbol{j}^e = 0$, where the electric current density is $j^e_i = \sigma_{ij} E_j$, one gets

$$0 = \partial_t (\boldsymbol{\nabla} \cdot \boldsymbol{D}) + \boldsymbol{\nabla} \cdot \boldsymbol{j}^e \, . \tag{7}$$

Equations (6) and (7) describe so far the full nonlinear dynamics of the problem. Since Eq.(7) gets rather complicated we expand it for the static case $\partial_t \phi = 0$ only and obtain

$$0 = 2\frac{\sigma_a}{\sigma_\perp} \sin\theta \cos\theta \left(V - \partial_z \phi(z)\right) \partial_z \theta - \left(1 + \frac{\sigma_a}{\sigma_\perp} \sin^2\theta\right) \partial_z^2 \phi \quad . \tag{8}$$

Linear stability analysis and a standard weakly nonlinear calculation reveals [Zimmermann et al 1996] that there is a new instability branch due to the polarization. This branch can, for certain parameter values, collapse and the trivial undisturbed solution becomes stable again (restabilization). To get an overview of what could happen for large director deformations we have integrated the coupled system of Eqs.(6,8) numerically. As a characterization of the nonlinear director field the integrated director deformation is introduced

$$B = \frac{1}{2} \int_{-\pi/2}^{\pi/2} \theta(z) \, dz \tag{9}$$

and interpreted as an order parameter. B vanishes for the basic state ($\theta_g = 0$). For the new instability branch the bifurcation behavior is complex. In the range $p_0 V_c < 0$ and $F < 0$ the elastic, dielectric and the ferroelectric torques favor different equilibrium states $\theta = 0$, $\theta = 0$ or π, and $\theta = \pi$, respectively. Since the dielectric torque dominates at very high voltages ($\eta \to \infty$) two (linear) stable equilibrium states are possible with $B = 0$ or $B \to \pi^2/2$, respectively. In the latter state (π-state) one has $\theta(z=0) \approx \pi$ at the cell center, while θ bends back to $\theta(z = \pm\pi/2) = 0$ near the boundaries.

For intermediate voltages the bifurcation diagrams are plotted as functions $B(\eta)$ in Fig.1 with the reduced voltage $\eta = \frac{V - V_c}{V_c}$.

Motivated by the amplitude equation [Zimmermann et al 1996] the numerical studies were done here for three distinct values of F_{11}/F_{22} belonging to the three regimes called I, II, and III, which describe the three possibilities of the Frederiks instability and the restabilization to be continuous or discontinuous.

In regime I the bifurcation at $\eta = 0$ is supercritical while the restabilization at η_r is also continuous (inverted backward). This restabilization threshold is indeed reached, since after increasing first with increasing values of η, B then decreases back to $B = 0$. Somewhat above η_r there is an unstable solution between the restabilized ground state and the isolated π-state. On the other hand, in regimes II and III the branch that bifurcates from the ground state ($B = 0$) at $\eta = 0$ never comes back to $B = 0$, i.e. the restabilized ground state is generally not reached in experiments. Above η_r there is an unstable branch between the ground state and the π-state. In regime III the

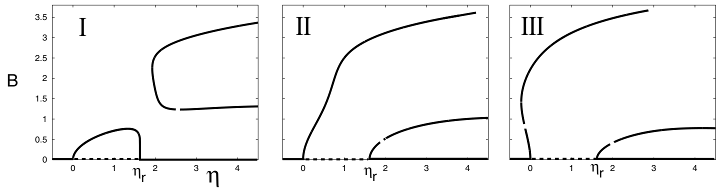

Fig. 1. *The integrated director deformation B as function of the reduced voltage $\eta = (V - V_c)/V_c$ for $F = -0.2$ at 3 different values of F_{11}/F_{22} ($= 2.9, 1.5, 0.5$ from left to right).*

bifurcation at $\eta = 0$ is subcritical showing a hysteresis, which can be quite large, especially for small values of F_{11}/F_{22}.

It should be mentioned, however, that the transition from the restabilization scenario to the π-state scenario does not necessarily take place at the same parameters, where regime I switches to regime II.

3.2 Electroconvection

In addition to the general Frederiks transition presented here, also electroconvection can occur, when an external electric field is applied to the film [Ried et al 1996]. While the Frederiks transition is homogeneous in x-direction ($q_c = 0$), electroconvection shows a pattern along x with a characteristic wavenumber $q_c \neq 0$. For a certain range of material parameters both instability types are possible and a competition between Frederiks and electroconvective instability takes place. If a pure electroconvective pattern formation is searched, we suggest experiments with $F < -1/4$, since no Frederiks transition is possible there. For DC voltages, in addition, all parameter values $F < 0$ will lead to an electroconvective transition, if the field is parallel to the polarization. On the other hand all (big) positive values of F will prefer the Frederiks transition $q_c = 0$ (cf. [Zimmermann et al 1996] for details).

In nematics there are two regimes for AC voltage electroconvection, one at low frequencies (conductive regime) and one at high frequencies (dielectric regime), with different thresholds and critical wavelengths for the cellular convection pattern. These known results [de Gennes and Prost 1993] are qualitatively unaffected by the presence of both, fixed layers and the macroscopic polarization in smectics C* for low and high frequencies of the applied electric AC field. At intermediate frequencies, however, a new "subharmonic regime" appears as the first unstable mode in Sm C* [Ried et al 1996]. Its threshold voltage increases with decreasing polarization. For vanishing polarization this regime does not exist and is therefore not accessible in other liquid crystal phases such as nematics or smectics C. Under certain conditions a codimension-3 point is found, where the three different instabilities (conductive, dielectric and subharmonic, all with different wavelengths and different temporal behavior) compete at onset.

4 Layer and In-Plane Instabilities Combined

If the Frederiks transition and the electroconvective instability in smectic C or C* is treated more realistically, i.e. if the assumption of fixed layers has been relaxed, it is obvious that layer undulations come into play. Not only is there a competition between Frederiks, electroconvection and electrobuckling instability, all of these three distinct instability types will acquire some features of the others showing flow, layer undulation and static director reorientation. Work on special aspects of such systems is in progress.

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note 1: This is a thermodynamic instability, where the positional order breaks down on very large length scales due to the divergence of undulational fluctuations, cf. [de Gennes and Prost 1993].

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note 2: In the thermal case the mechanism is more complicated involving dissipative processes as well.

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