Polarization Frederiks-Transition in SmC* Liquid Crystal Films

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Abstract

A new Frederiks-transition is predicted for freely suspended SmC*-liquid crystal films. Frederiks transitions in nematic liquid crystals are the result of the competition between two orientational effects. One coming from the orientation of the director field at the boundary and one from the orientational effect due to an external electrical or magnetic field acting on the bulk orientation of the director field via the magnetic or dielectric susceptibility anisotropy. Here we discuss the consequences of the additional ferroelectric torques. A new branch for the splay Frederiks transition due to the presence of the spontaneous polarization in SmC*-liquid crystal films is predicted, and it is pointed out that on this new branch a transition from supercritical to subcritical bifurcation behavior can occur.

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I. Introduction: External field induced orientational transitions in liquid crystals, especially in nematic liquid crystals have a long history and have been observed for the first time by Frederiks and Repiewa [1]. During the last two decades those orientational instabilities experienced technologically important applications in todays modern liquid crystal displays [2]. Here a new orientational transition occurring in smectic C* liquid crystals films is investigated.

The smectic phases are organized in layers, where in the smectic C phase (SmC) the director **n** is tilted by a fixed angle ψ relative to the layer normal **k**. The projection of **n** onto the plane of the smectic layers is the **c** director, which can be observed by polarized light normal to the layer. Due to the existence of **k** and **c** this phase is biaxial.

The chiral smectic C* phase (SmC*) shows in addition an intrinsic twist of the director from layer to layer. This additional symmetry breaking ($C_{2h} \rightsquigarrow C_2$ locally) allows microscopic electric dipoles to form a spontaneous electric polarization **P**, which lies in the smectic planes (perpendicular to both **k** and **c**) and is twisted, too. We will neglect this twist in the following thus assuming that the thickness of the freely suspended film is small compared to the pitch of the helielectric C* phase [3], which is typically $\sim 1 \ldots 10\,\mu m$.

Thus we consider as the ground state (without applied external fields) a homogeneous structure, where each, **P** and **c**, have on average one specific preferred direction, perpendicular to each other. Since we assume the layers to be rigid, we can use a 2-dimensional model to describe the system. The hydrodynamic degree of freedom is a rigid rotation of the orthogonal vectors **P** and **c** in the layer plane denoted by the angle θ (cf. Fig.1). The in-plane spontaneous polarization **P** is always perpendicular to the **c** director,

$$\mathbf{P} = p_0\,(-\sin\theta, 0, \cos\theta)\,, \qquad \mathbf{c} = (\cos\theta, 0, \sin\theta)\;, \tag{1}$$

where the angle θ describes the orientation within the film plane. Without external fields the ground state is homogeneous with $\theta_g = 0$ and $\theta_g = \pi/2$ for planar and homeotropic boundary conditions, respectively.

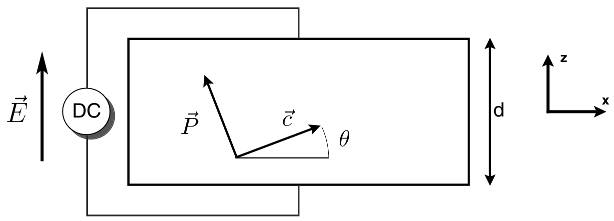

Figure 1: *One layer of SmC* is shown from above. The electric field is applied along the z-direction. The length of the film (in x-direction) is assumed to be much longer than its width d. The layer normal* **k** *is perpendicular to the plane of drawing (cf. [4]).*

II. Torque Balance: An applied electric field has an orientational effect on the director due to the dielectric anisotropy, ε_a. If ε_a is positive (negative), a parallel (perpendicular) alignment of the director with the external field is energetically favorable and the associated torques on the director are quadratic in the external field amplitude (or voltage). The presence of a spontaneous polarization in SmC* layers leads to an additional, ferroelectric torque, since **P** wants to be aligned parallel to the external field. On the other hand **P** is rigidly coupled to **c** giving a torque on **c**, which is linear in the field amplitude. Having fixed the director orientation at the boundaries (in z-direction) an external field can thus lead to an inhomogeneous texture (a z-dependent director field **c**). Then, of course, orientational elastic torques, characterized by Frank constants, will occur that will try to restore the homogeneous director orientation.

We investigate static director deformations above a DC-driven splay Frederiks-transition, which are assumed to be homogeneous in x-direction. The backflow is very weak in this situation [5] and will be neglected. For possible back flow effects cf. [6]. With the notation of Refs. [7] and using the following scalings for space, $x = x'd/\pi$, voltage, $V = V'\pi(F_{22}/\varepsilon_\perp)^{1/2}$, time, $t = t'\tau_0 = t'(\gamma_1 d^2/F_{22}\pi^2)$, and polarization, $p_0' = p_0(d/\pi)(\varepsilon_\perp F_{22})^{-1/2}$, the nonlinear balance equation of the director takes the form

$$\partial_t\theta = \Big(\frac{F_{11}}{F_{22}}\,\sin^2\theta + \cos^2\theta\Big)\,\partial_z^2\,\theta + \Big[\Big(\frac{F_{11}}{F_{22}} - 1\Big)\Big(\partial_z\theta\Big)^2 + \frac{\varepsilon_a}{\varepsilon_\perp}\,V^2\Big]\,\cos\theta\,\sin\theta - p_0\,V\,\sin\theta, \tag{2}$$

where γ_1 is the rotational viscosity, $\varepsilon_\perp$ the dielectric constant perpendicular to the director **c**, and F_{22} the splay and F_{11} the bend elastic constant introduced first in [8]. Eq.(2) can also be obtained using equations from [9]. In writing down Eq.(2) flexoelectric contributions have been discarded.

III. DC-Thresholds: Threshold values for applied DC voltages at which the Frederiks transition occurs are calculated from the linear part of Eq.(2) with the ansatz, $\theta(t,z) = e^{\lambda t} A_0 \cos z$, for deviations θ from the planar ground state ($\theta_g = 0$). The basic state becomes unstable for positive growth rates λ and the threshold is defined by the condition $\lambda = 0$. For SmC films and nematics ($p_0 = 0$) the threshold (critical) voltage is given by

$$V_c^2 = \varepsilon_\perp / \varepsilon_a , \tag{3}$$

which shows that the threshold is independent of the sign of the applied electric field **E** (or voltage) and exists only for finite positive values of ε_a. This behavior is changed qualitatively in smectic C^* liquid crystals due to the presence of the polarization **P**, since **E** parallel to **P** ($p_0 V > 0$) is energetically preferred compared to the case **E** antiparallel to **P** ($p_0 V < 0$). Thus, even for vanishing dielectric anisotropy, $\varepsilon_a = 0$, one finds an orientational transition of the Frederiks type ("polarization Frederiks effect") with the threshold [6]

$$V_c = -1/p_0 \, . \tag{4}$$

This instability only occurs, if the spontaneous polarization and the applied electrical field are antiparallel to each other ($p_0 V < 0$) in the ground state, leading to a torque on **P** (and **c**) for any deviation from the ground state.

In the general case, i.e. for finite values of p_0 and ε_a, both orientational torques are present and the threshold formula of the general Frederiks instability due to the applied DC voltage is

$$p_0 V_c = \frac{1}{2F}\left[1 \pm \sqrt{1+4F}\,\right], \qquad \text{with} \qquad F \equiv \frac{\varepsilon_a}{\varepsilon_\perp p_0^2} \, . \tag{5}$$

This formula contains both, the traditional splay Frederiks effect (due to dielectric torques) and the pure "polarization Frederiks effect" (due to ferroelectric torques), Since polarization and director are rigidly coupled, both effects can either enhance each other (for $p_0 V < 0$ and $\varepsilon_a > 0$) and therefore reduce the threshold voltage, or counteract each other (for $p_0 V > 0$ and $\varepsilon_a > 0$) increasing the threshold voltage (cf. Fig.2).

In contrast to the traditional splay Frederiks transition that exists only for $\varepsilon_a > 0$, the general Frederiks transition can exist even for $\varepsilon_a \leq 0$, if $p_0 V < 0$ (external field antiparallel to the polarization) and if p_0 exceeds a critical value $p_0 > p_c$, where

$$p_c = \sqrt{\frac{-4\varepsilon_a}{\varepsilon_\perp}} \tag{6}$$

(i.e. if $F > -1/4$). In that case the destabilizing effect due to the polarization overcomes the stabilizing effect due to the dielectric anisotropy. This range is indicated in Fig.2, where below the solid line the planar initial configuration is Frederiks-unstable. This branch is new and owes its existence to the polarization Frederiks effect. However, since the destabilizing effect is linear in the external field strength while the stabilizing one is quadratic, the latter will win above a certain (higher) threshold, above which the ground state becomes stable again. This restabilization is shown as dotted curve in Fig.2. Since the stabilizing effect vanishes for $\varepsilon_a \to 0_-$, the dotted curve diverges in this limit. On the other hand, if p_0 is too small ($p_0 < p_c$), the linear destabilizing effect is too weak to trigger the instability and the ground state is Frederiks stable. The dashed line in Fig.2 represents the threshold of the traditional Frederiks instability ($\varepsilon_a > 0$) in the presence of finite p_0. In the limit of vanishing polarization and positive dielectric anisotropy ($F \to +\infty$) the two instability lines coalesce restoring the degeneracy due to the $\pm V$ symmetry in this limit.

IV. Weakly Nonlinear Behavior: The traditional splay Frederiks transition driven by the dielectric torque is always continuous (supercritical). To check the bifurcation behavior for the general case ($p_0 \neq 0$), we expand Eq.(2) around the critical value V_c, with the expansion parameter

$$\eta = \frac{V - V_c}{V_c} \ . \tag{7}$$

Additionally we consider only small amplitudes of the linear mode $\theta(t, z) = A(t)\cos z$ with $A \propto \eta^{1/2}$. In leading order the cubic Landau equation

$$\tau_0\, \partial_t A = (\eta - g A^2)\, A, \tag{8}$$

is obtained from Eq.(2), where τ_0 is the relaxation time of the director and the sign of the nonlinear coefficient g determines the bifurcation type:

$$\tau_0 = \frac{2\,F}{1 + 4F \pm \sqrt{1 + 4F}} \ , \qquad\qquad g = \frac{3}{8}\,\frac{1 + \frac{8\,F_{11}}{3\,F_{22}}F \pm \sqrt{1+4F}}{1 + 4F \pm \sqrt{1+4F}} \ . \tag{9}$$

The $\pm$ signs correspond to the $\pm$ signs in Eq.(5) and to the two branches in Fig.2. The nonlinear coefficient is always positive (supercritical bifurcation) for the dashed branch in Fig.2 (upper sign), whereas the general Frederiks transition for $p_0 V_c < 0$ (lower sign, solid line in Fig.2) can also be discontinuous (subcritical), depending on the elastic constants, the dielectric anisotropy and the polarization. The line, where the nonlinear coefficient is zero and which separates super- and subcritical regimes, is plotted in Fig.3.

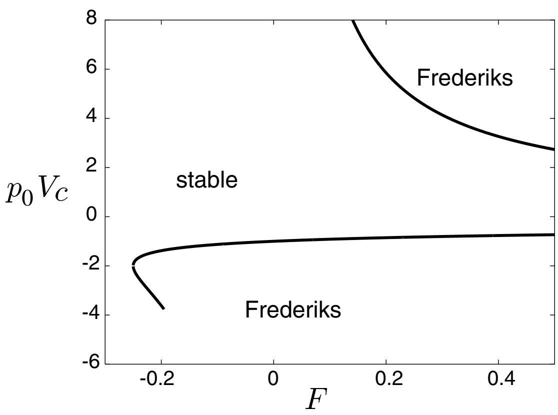

Figure 2: *The threshold for the general splay Frederiks transition is shown as function of the quantity $F = \varepsilon_a/(\varepsilon_\perp p_0^2)$. The dashed curve branches out of the traditional Frederiks transition and the solid line describes the new branch due to the polarization Frederiks effect, whereas the dotted line shows the linear restabilization of the basic state $\theta_g = 0$.*

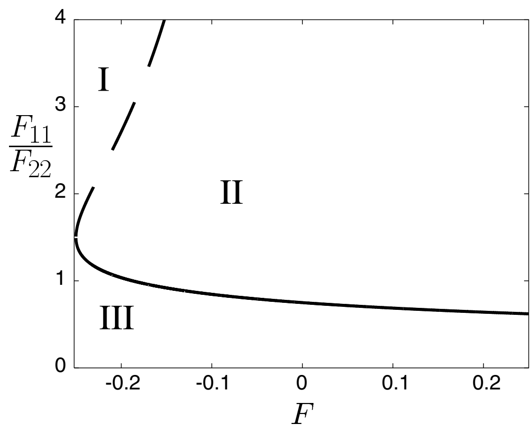

Figure 3: *The zeros of the nonlinear coefficient g (cf. Eq.(8)) are given for $p_o V_c < 0$ in the parameter space F_{11}/F_{22} vs. F. Above and below the solid (dotted) line the polarization Frederiks transition (the restabilization) is continuous and discontinuous, respectively.*

V. Nonlinear Regime: The static director deformation and therefore the bifurcation behavior beyond the validity range of the amplitude equation (8), e.g. for large director deformations, has been determined by solving the static part of Eq.(2) numerically. As a characterization of the nonlinear director field the integrated director deformation is introduced

$$B = \frac{1}{2} \int_{-\pi/2}^{\pi/2} \theta(z)\, dz \qquad (10)$$

and interpreted as an order parameter. B vanishes for the basic state ($\theta_g = 0$) and reduces to $B = A$ for small director deformations $\theta(z) = A \cos z$. Above the dashed threshold line in Fig.2 ($p_0 V_c > 0$ and $F > 0$) the bifurcation from the basic state is continuous and the dielectric torque dominates both, the ferroelectric and the elastic one. Thus, $\theta = \pi/2$ is favored as equilibrium state and for large voltages, $\eta \to \infty$, B approaches $\pi^2/4$ from below.

For the new instability branch (solid line in Fig.2) the bifurcation behavior is more complex. In the range $p_0 V_c < 0$ and $F < 0$ the elastic, dielectric and the ferroelectric torques favor different equilibrium states $\theta_g = 0$, $\theta = 0$ 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

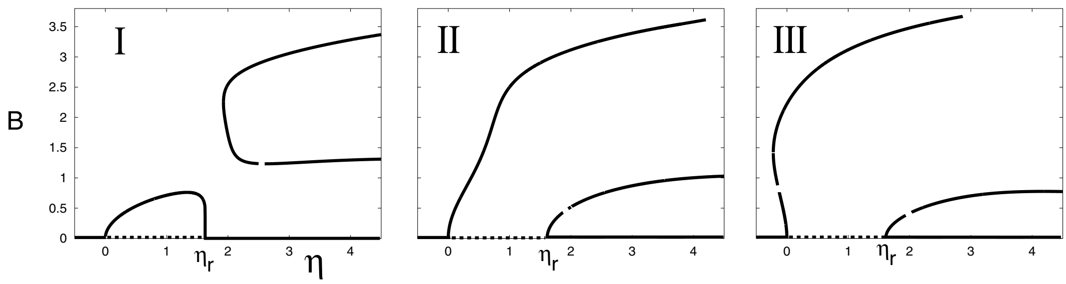

Figure 4: *The integrated director deformation B, introduced in Eq.(10), is plotted as function of the reduced voltage $\eta = (V - V_c)/V_c$ for the three regimes I, II and III (cf. Fig.3) with $F = -0.2$, where linear restabilization of the basic state occurs above threshold. The dotted lines correspond to the unstable solutions and the solid lines to stable ones (with $F_{11}/F_{22} = 2.9,\ 1.5,\ 0.5$ in regime I, II and III respectively).*

in Fig.4. In regime I (cf. Fig.3) the bifurcation has a rather complex structure. At $\eta = 0$ the bifurcation is supercritical. With increasing values of η also B increases, then however, generally bends back to $B = 0$, where the restabilization threshold is reached at $\eta_r = 1.53$. (Only for F_{11}/F_{22} values very close to regime II the branch leading to the restabilization breaks apart and the two stable branches at the upper left in Fig.4 (I) coalesce, thus prohibiting restabilization.) Beyond η_r there is an unstable solution between the restabilized ground state and the π-state. Similar pictures arise for regimes II and III above the linear restabilization. However, in these regimes 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. In regime III the bifurcation at $\eta = 0$ is subcritical and there are two linear stable (solid lines) and one linear unstable (dashed) branch immediately below $\eta = 0$. The range of this hysteresis, measured by η_{SN}, can be quite large, especially for small values of F_{11}/F_{22}.

VI. Discussion and Conclusion: For homeotropic orientation of the director the polarization is perpendicular to the applied electric field. Thus, there is no threshold for a Frederiks transition and any non-zero external field, even infinitesimally small, leads to a ferroelectric torque and to a distortion of the ground state. The reason for this difference to the planar case is that $\mathbf{P} \perp \mathbf{E}$ is no equilibrium state, while $\mathbf{P} \parallel \mathbf{E}$ (parallel or antiparallel) is either a stable or an unstable equilibrium state. The dielectric torques behave differently, since both cases, $\mathbf{c} \parallel \mathbf{E}$ and $\mathbf{c} \perp \mathbf{E}$ are (stable or unstable) equilibrium states (depending on the sign of ε_a). Therefore, in systems without a polarization, the planar and homeotropic cases are similar to each other and both show a finite threshold for Frederiks transitions in contrast to SmC* films.

For external AC voltages the ferroelectric torque, being linear in the field strength, is almost averaged to zero during a full cycle. Thus, for AC frequencies larger than the reciprocal reorientation time of the director the ferroelectric torque is small, while for lower frequencies a statistical description is necessary. Of course, the dielectric torque, which is quadratic in the field strength is not averaged out in an AC field. Thus, for AC fields the Frederiks transition in SmC* films is qualitatively similar to that in SmC films or nematics and does not carry new features. For a finite wavelength electroconvective instability, which is the preferred instability type for strong negative values of F, the situation is different for AC-driving fields, where the existence of the polarization leads to a qualitatively new subharmonic bifurcation [10].

The experimental detection of the very special effects of the polarization in the Frederiks transition of a planar ground state and external DC field, discussed in the previous sections, should be no problem using substances of sufficiently high purity.

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