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Threshold voltage of the Freedericksz transition in a hexatic smectic I phase†

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Using the linearized equations of the elastic continuum theory, simple formulae for the threshold voltage for the Freedericksz transition in planar films of the hexatic smectic I phase are derived. In contrast to the Freedericksz transition of a nematic film, this threshold strongly depends on the film thickness. The behaviour of the film in the weakly non-linear region slightly above the threshold is also considered to characterize the film instability.

1. Introduction

Most investigations of the Freedericksz transition refer to thin films of nematic liquid crystals [1]. Applying an electric field to a suitably prepared planar or homeotropic film, elastic distortions appear for a voltage above a certain threshold value. It was found both experimentally and theoretically that the threshold voltage does not depend on the film thickness. Freedericksz transitions also occur in films of smectic C liquid crystals [2]. Although the threshold condition for the instability turns out to be more complicated due to the occurrence of discontinuous transitions [3, 4], the conventional elastic theory predicts that threshold voltages should not depend on the sample thickness.

Tilted hexatic liquid crystals, however, which exhibit a bond orientational order [5], are expected to behave in a different manner. Recently, Gingras [6] has performed numerical calculations for a smectic I liquid crystal film subjected to a magnetic field. It was found that the threshold behaviour is rather complicated for a hexatic film with smectic layers oriented parallel to the substrates. Besides the continuous Freedericksz transition, another transition mainly caused by a reorientation of the bonds was also detected.

In this paper, the threshold of the film instability for planar films of the hexatic smectic I phase is obtained analytically by performing a linear stability analysis. An investigation of the weakly non-linear region just above the threshold voltage is added in order to decide whether the Freedericksz transition is continuous or discontinuous.

Figure 1 depicts the molecular ordering in the layers of a smectic I liquid crystal. Although the triangular lattice in the smectic layers is molten, a long-range bond orientational order still exists. This order is associated with the orientation of the lines connecting the centres of gravity of neighbouring molecules. That means that the local hexagon formed by the six nearest neighbours of a particle is oriented macroscopically in an undisturbed sample. If distortions occur, the hexagon can be rotated by an angle η. The preferred direction of the molecular long axes (director **n**) and the normal to the smectic layers enclose a fixed polar angle θ, so that the director projection on the layer plane is oriented towards a vertex of the hexagon. Director rotations due to distortions in the sample are defined by the azimuthal angle ϕ. In figure 2, the director angle ϕ and the bond angle η characterize deviations from the perfect planar alignment of a smectic I film confined between two boundary plates. Supposing a strong anchoring of the director and the bond direction, the angle ϕ and η are equal to zero at the substrates. The direction of the smectic layers is not necessarily parallel to the film normal, but can enclose a non-zero

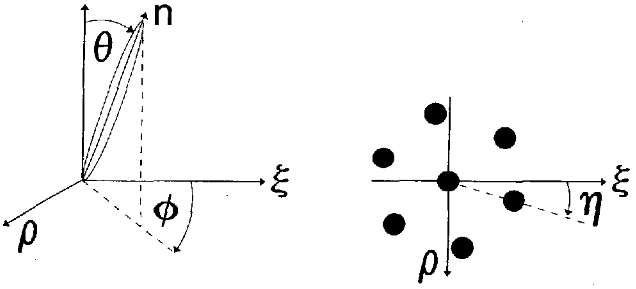

Figure 1. Degrees of freedom necessary to describe the distortions in a hexatic smectic I phase [7]. The coordinate axes ρ and ξ are parallel to a smectic layer. Rotations of the director **n** are characterized by the azimuthal angle ϕ. The angle η defines the rotation of the hexagon formed by the six nearest neighbours of a particle.

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† Dedicated to Professor A. Saupe on the occasion of his 70th birthday.

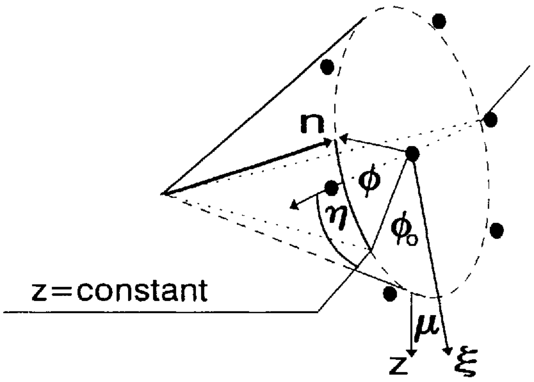

Figure 2. Reorientation in a planar film of the smectic I phase. The angles ϕ and η define the rotation of the director **n** and the hexagon of the bonds, respectively. The axis z is perpendicular to the film interfaces, whereas the axis ξ lies on the smectic plane. Deviations from the bookshelf geometry are characterized by a non-zero layer tilt angle μ.

angle μ. The symmetrical configuration with $\mu = 0$ is the called bookshelf geometry.

Theoretical investigations suggest [7] that in a smectic C phase a weak hexatic order should also exist as a consequence of the coupling between the bond orientational order and the molecular tilt. In this case, the results obtained below also provide an extension of the conventional theory for the Freedericksz transition in planar films of the smectic C phase.

A film of a smectic I liquid crystal with thickness d has the free elastic energy [6]

$$F_1 = \frac{1}{2}\int_{-d/2}^{+d/2} \mathrm{d}z[K(\nabla\phi)^2 + L(\nabla\eta)^2 + 2G\nabla\phi\nabla\eta] \qquad (1)$$

where the anisotropy of the elastic behaviour is neglected. Since the elastic free energy must be positive by definition, the elastic constants for director and bond distortions obey the conditions $K > 0$, $L > 0$ and $KL - G^2 > 0$.

In the case of smectic layers with a tilt angle μ, the free electric field energy is represented [4] by

$$F_2 = -\frac{1}{2}\int_{d/2}^{+d/2} \mathrm{d}z\Delta\varepsilon E^2 \sin^2\theta\cos^2\mu(\cos(\phi + \phi_o) - \cos\phi_o)^2 \qquad (2)$$

where $\Delta\varepsilon > 0$ and ϕ_o is defined in figure 1. As the dielectric anisotropy $\Delta\varepsilon = \varepsilon_{II} - \varepsilon_\perp$ is supposed to be small ($\Delta\varepsilon\sin^2\theta \ll \varepsilon_\perp$), the electric field E is approximately constant within a distorted sample. Here ε_{II} and $\varepsilon_\perp$ are the dielectric susceptibilities measured parallel and perpendicular to the director, respectively. Finally, the free energy contribution [5]

$$F_3 = -\frac{1}{36}h\int_{-d/2}^{+d/2} \mathrm{d}z\cos 6(\phi - \eta) \qquad (3)$$

(with $h > 0$) provides six preferred directions for the director due to the hexatic order of the bonds. For an equilibrium state of the film, the total free energy $F = F_1 + F_2 + F_3$ is a minimum. If deviations from the equilibrium state occur, the dynamics of the director and the bond angle are described by the phenomenological equations

$$\left.\begin{aligned} -\Gamma_1\frac{\partial\phi}{\partial t} &= \frac{\delta F}{\delta\phi} \\ \text{and} \qquad & \\ -\Gamma_2\frac{\partial\eta}{\partial t} &= \frac{\delta F}{\delta\eta}, \end{aligned}\right\} \qquad (4)$$

where the conditions

$$\left.\begin{aligned} \phi(z = -d/2) = \phi(z = +d/2) &= 0 \\ \text{and} \qquad & \\ \eta(z = -d/2) = \eta(z = +d/2) &= 0 \end{aligned}\right\} \qquad (5)$$

are satisfied at the boundaries.

2. Instabilities in the case of the bookshelf geometry

2.1. *Threshold voltage*

For the case that the smectic layers are oriented perpendicular to the boundary plates, we insert $\mu = 0$ and $\phi_o = \pi/2$ in integral (2). Thus F_2 is simplified to

$$F_2 = -\frac{1}{2}\int_{-d/2}^{+d/2} \mathrm{d}z\,\Delta\varepsilon E^2\sin^2\theta\sin^2\phi. \qquad (6)$$

The equations of motion (4) are written explicitly

$$\left.\begin{aligned} &\Gamma_1\phi_t = K\phi_{zz} + G\eta_{zz} + b\cos\phi\sin\phi - \tfrac{1}{6}h\sin 6(\phi - \eta) \\ &\text{and} \\ &\Gamma_2\eta_t = G\phi_{zz} + L\eta_{zz} + \tfrac{1}{6}h\sin 6(\phi - \eta) \end{aligned}\right\} \qquad (7)$$

with the notation $b = \Delta\varepsilon E^2\sin^2\theta$.

Linearizing (7) we obtain

$$\left.\begin{aligned} \Gamma_1\phi_t &= K\phi_{zz} + G\eta_{zz} + b\phi - h(\phi - \eta) \\ \text{and} \qquad & \\ \Gamma_2\eta_t &= G\phi_{zz} + L\eta_{zz} + h(\phi - \eta) \end{aligned}\right\} \qquad (8)$$

The system (8) of linear differential equations can be solved by using an ansatz

$$\begin{pmatrix}\phi\\ \eta\end{pmatrix} = \begin{pmatrix}\phi_o\\ \eta_o\end{pmatrix}\exp(ikz + \lambda t) \qquad (9)$$

where $i = \sqrt{-1}$. Then two linear algebraic equations

$$\left.\begin{aligned} (-Kk^2 + b - h - \lambda\Gamma_1)\phi_o - (Gk^2 - h)\eta_o &= 0 \\ \text{and} \qquad & \\ (Gk^2 - h)\phi_o + (Lk^2 + h + \lambda\Gamma_2)\eta_o &= 0 \end{aligned}\right\} \qquad (10)$$

result for ϕ_o and η_o. The system (10) can only be solved in those cases where its determinant is equal to zero. This condition leads to the dispersion relation

$$\Gamma_1\Gamma_2\lambda^2 - [\Gamma_2(-Kk^2+b-h) - \Gamma_1(Lk^2+h)]\lambda \\ -(-Kk^2+b-h)(Lk^2+h) - (Gk^2-h)^2 = 0 \quad (11)$$

connecting the rate λ of growing or decaying deformations with k. According to (9), neutral stability is achieved if $\lambda = 0$. Then k satisfies the equation

$$(-Kk^2+b-h)(Lk^2+h) + (Gk^2-h)^2 = 0, \quad (12)$$

which reduces to a quadratic equation for k^2. Equation (12) has two real solutions, $\pm k_1$ and two purely imaginary solutions, $\pm ik_2$ (with real k_2). A general solution of the linearized eigenvalue problem (8) with boundary conditions (5) is a linear combination of the functions $\cos k_1z$ and $\cosh k_2z$. The coefficients of this linear combination are connected by the equations (10), which are linear dependent as a consequence of the relation (12). Thus we choose the second equation and obtain

$$\begin{pmatrix}\phi\\ \eta\end{pmatrix} = A\begin{pmatrix}1\\ R_1(k_1)\end{pmatrix}\cos k_1z + B\begin{pmatrix}1\\ R_2(k_2)\end{pmatrix}\cosh k_2z \quad (13)$$

where

$$R_1(k_1) = \frac{-Gk_1^2+h}{Lk_1^2+h}$$

and

$$R_2(k_2) = \frac{Gk_2^2+h}{-Lk_2^2+h}.$$

It can be proved that the boundary conditions (5) are satisfied if

$$\cos(k_1d/2)[R_2(k_2) - R_1(k_1)] = 0. \quad (14)$$

Since k_1 and k_2 are real, the only possible solutions of equation (14) are found to be

$$k_1 = \frac{\pi}{d} + n\frac{2\pi}{d} \quad (15)$$

with integer n and the implication $B = 0$. According to equation (12), the bifurcation parameter b has the critical value

$$b_c = h + Kk_1^2 - \frac{(Gk_1^2-h)^2}{Lk_1^2+h}. \quad (16)$$

The lowest value of the instability threshold b_c is accompanied with $n = 0$ ($k_1 = \pi/d$), and the threshold voltage $U_c = E_cd$ satisfies the equation

$$U_c^2 = \frac{\pi^2K}{\Delta\varepsilon\sin^2\theta}\left[1 + \left(\frac{hd^2}{\pi^2K}\right) - \frac{\left(\dfrac{G}{K} - \dfrac{hd^2}{\pi^2K}\right)^2}{\dfrac{L}{K} + \dfrac{hd^2}{\pi^2K}}\quad.\right] \quad (17)$$

Let us consider two borderline cases. If the sample thickness d is small, so that $hd^2/\pi^2 \ll L, G$, then formula (17) leads to

$$\left(\frac{U_c}{U_o}\right)^2 = \left(1 - \frac{G^2}{KL}\right) + \left(1 + \frac{G}{L}\right)^2\left(\frac{d}{d_o}\right)^2 + O((d/d_o)^4) \quad (18)$$

where

$$U_o = \left(\frac{\pi^2K}{\Delta\varepsilon\sin^2\theta}\right)^{1/2}$$

is the threshold voltage for the Freedericksz transition of a smectic C film without bond orientation order ($h = G = 0$) and

$$d_o = \left(\frac{\pi^2K}{h}\right)^{1/2}$$

is a characteristic length. Otherwise, for a rather thick sample with $hd^2/\pi^2 \gg L, G$, the threshold voltage (16) becomes asymptotically

$$\left(\frac{U_c}{U_o}\right)^2 = 1 + \frac{L+2G}{K} - \frac{(L+G)^2}{K^2}\left(\frac{d_o}{d}\right)^2 + O((d_o/d)^4). \quad (19)$$

The plot $(U_c/U_o)^2$ versus $(d/d_o)^2$ in figure 3 demonstrates how the threshold voltage depends on the sample thickness.

2.2. *Behaviour slightly above the threshold voltage*

Now the non-linearity of equations (7) are taken into account in deciding whether the field induced transition is continuous or discontinuous. Using a perturbation procedure, the bifurcation type can be predicted by analysing the weakly non-linear region above the threshold voltage. For this purpose the trigonometric functions in equations (7) are expanded in a series up to terms of third order with respect to ϕ and η:

$$\left.\begin{aligned} K\phi_{zz} + G\eta_{zz} + b_c\phi - h(\phi-\eta) &= -(b-b_c)\phi \\ &\quad + \tfrac{2}{3}b_c\phi^3 - 6h(\phi-\eta)^3 + \Gamma_1\phi_t \\ \text{and}\qquad & \\ G\phi_{zz} + L\eta_{zz} + h(\phi-\eta) &= 6h(\phi-\eta)^3 + \Gamma_2\eta_t. \end{aligned}\right\} \quad (20)$$

These equations can be written as

$$D\begin{pmatrix}\phi\\ \eta\end{pmatrix} = \begin{pmatrix}f_1\\ f_2\end{pmatrix} \quad (21)$$

where D is a linear differential operator. Taking into account the boundary conditions (5), it is checked easily that D is a self-adjoint operator, and as the equation $D\psi = 0$ has the non-trivial solution

$$\begin{pmatrix}\psi_1\\ \psi_2\end{pmatrix} = \begin{pmatrix}1\\ R_1(k_1 = \pi/d)\end{pmatrix}\cos\frac{\pi z}{d}, \quad (22)$$

D is singular. According to Freedholm's alternative [9], equation (20) may be solved only in those cases where the condition

$$\int_{-d/2}^{+d/2} dz(f_1\psi_1 + f_2\psi_2) = 0 \tag{23}$$

is satisfied. Inserting the distortion angles

$$\left.\begin{aligned} \phi(z,t) &= A\cos\frac{\pi z}{d} \\ \text{and} \qquad \eta(z,t) &= A\,\frac{-Gk_1^2+h}{Lk_1^2+h}\cos\frac{\pi z}{d} \end{aligned}\right\} \tag{24}$$

in the expressions for f_1 and f_2 the condition (23) immediately leads to the non-linear equation

$$\Gamma\frac{dA}{dt} = (b-b_c)A - pA^3 \tag{25}$$

for the distortion amplitude A, where the coefficients are defined by

$$p = \tfrac{1}{2}b_c - \tfrac{9}{2}(1-R_1(k_1))^4h, \tag{26}$$

$$\Gamma = \Gamma_1 + R_1(k_1)^2\Gamma_2, \tag{27}$$

and $k_1 = \pi/d$. If p is positive, the transition to the distorted state is a forward bifurcation and distortions grow continuously with increasing voltage. After applying a voltage, which is a little bit higher than the threshold, a small initial distortion A_0 grows exponentially

$$A(t) = A_0\exp(t/\tau) \tag{28}$$

with the rise time $\tau = \Gamma/(b-b_c)$. According to equation (25) the amplitude of the distortions tends to

$$A = \pm\left(\frac{b-b_c}{p}\right)^{1/2} \tag{29}$$

for $t \gg \tau$. It should be emphasized that formula (29) is only valid in a region slightly above the threshold b_c. If p is negative, hysteresis is expected to occur and the time-dependent behaviour of the Freedericksz transition is more complicated.

3. Freedericksz threshold voltage in the case of tilted smectic layers

Let us briefly discuss some modifications of the results occurring if the smectic layers are not perpendicular to the bounding plates but enclose a non-zero angle μ. As previously, we assume that the director and the bonds are parallel to the substrate interfaces. The angle ϕ_o (see figure 2) is connected to the layer tilt μ by the relation $\cos\phi_o = \tan\mu/\tan\theta$. Now, instead of formula (16), the linear stability analysis leads to

$$\tilde{b}_c = \frac{1}{\sin^2\phi_o\cos^2\mu}\left[h + Kk_1^2 - \frac{(Gk_1^2-h)^2}{Lk_1^2+h}\right] \tag{30}$$

$(k_1 = \pi/d)$. Obviously, the instability threshold $\tilde{b}_c$ is increased in comparison to the case $\mu = 0$ and $\phi_o = \pm\pi/2$. The relations (24) for the deformations remain valid in the vicinity of the threshold, but (25) is modified to an equation of the type

$$\Gamma\frac{dA}{dt} = [(b-\tilde{b}_c)\sin^2\phi_o\cos^2\mu]A - \tilde{p}A^2 \tag{31}$$

for the deformation amplitude. This equation describes a trans-critical bifurcation, which is usually accompanied with hysteresis. Then the experimentally observed instability is expected to occur even at a threshold for b which is somewhat lower than the stability limit $\tilde{b}_c$, and the discontinuous (first-order) transition leads to a strongly distorted state [4].

4. Discussion

Let us restrict the discussion to a bookshelf configuration ($\mu = 0$), since the general conclusions are not influenced if the layer tilt angle μ is non-zero. Figure 3 depicts the limits of stability for planar films. In the region below each curve, the undistorted planar alignment is stable with respect to small fluctuations of ϕ and η. These curves also define the threshold of the film instability if hysteresis effects can be neglected.

In contrast to a nematic film, the critical voltage for the Freedericksz transition depends on the film thickness d. In figure 3, for several ratios L/K of the elastic constants, it is demonstrated how the threshold voltage depends on the film thickness. If the film thickness is small ($d \ll d_o$),

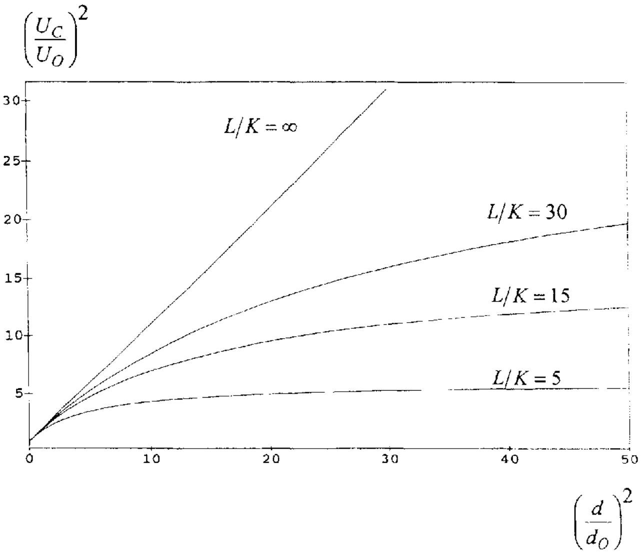

Figure 3. The square of the threshold voltage is plotted versus the square of the film thickness in dimensionless units for several ratios of the elastic constants L and K. The third elastic constant G is assumed to be zero; U_o and d_o are defined in the text.

the square of the threshold U_c^2 is described by a linear function of d^2. Then U_c^2 increases with growing d^2. For a thick sample ($d \gg d_o$), U_c^2 tends to a constant value. If the bond orientational order is pronounced, as is expected in a hexatic smectic I phase, deviations from a constant threshold voltage should be clearly observable. Probably, the coupling of the molecular tilt to the bonds also produces a weak bond orientational order in a smectic C phase. In this case, the thickness dependence of the threshold voltage should be observable if the sample thickness d is comparable to $d_o = \pi\sqrt{(K/h)}$ and if the elastic constant L is not small in comparison to K.

There could be a simple way of distinguishing between the behaviour of the hexatic smectic I phase and the crystal J phase, which is a true crystalline phase with a triangular lattice in each layer [5]. For a crystalline order, the elastic constant L is infinitely high. Inserting $L/K = \infty$ in equation (17), we obtain the formula $(U_c/U_o)^2 = 1 + (d/d_o)^2$. Accordingly, the threshold voltage of a film possessing crystalline ordering always increases with growing film thickness, whereas expression (19) tends to a constant value if $d/d_o \rightarrow \infty$.

It should be noted that the bifurcation analysis cannot exclude the existence of a further discontinuous transition to a strongly distorted state which does not branch from the initial state $\phi = \eta = 0$. Considering also strongly distorted states by a numerical calculation, Gingras [6] found a first order transition, which is mainly based on a reorientation of the bonds. For a certain range of the parameter h, this transition has a lower threshold than the Freedericksz threshold. Such a discontinuous transition is not detectable by the analytical approach presented, since the stability is only checked for relatively small deviations from the homogeneous film configuration.

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