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Phase diagrams of cholesteric films in electric fields

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Planar films of cholesteric liquid crystals exhibit several types of instabilities in electric fields. A periodic equilibrium structure can appear when the field is parallel to the helix axis. Conditions for the occurrence of the modulated phase have been derived by an analytical theory, which describes long wavelength distortions in the vicinity of Lifshitz points.

1. Introduction

A classical problem of liquid crystal physics is the director reorientation in thin nematic films subject to an external magnetic or electric field. The most familiar film instability is the Freedericksz transition of planar oriented nematic liquid crystals with positive dielectric anisotropy [1]. This transition starts at a definite threshold voltage and leads to a director rotation towards the field direction. The observed film textures below and above the threshold voltage are homogeneous, since distortion gradients in the film are perpendicular to the bounding plates. Recently, Lonberg and Meyer [2] have found both experimentally and theoretically that instead of the Freedericksz transition a periodic equilibrium structure visible as a striped texture appears, when the elastic anisotropy of the nematic material is large enough. Main-chain polymeric liquid crystals seem to be suitable for the formation of a periodic pattern.

Field-induced periodic distortions of cholesteric films have been well known for a long time. Hurault [3] obtained a formula for the threshold field strength of modulation in a cholesteric phase with the helix axis parallel to the applied field. Chigrinov *et al.* [4] have improved this theory by taking into account boundary conditions. They have found that there is a competition between the Freedericksz effect and the tendency of cholesteric films to form a modulated structure. If the ratio of the film thickness and the helix pitch is small a Freedericksz transition takes place. But for sufficiently large total twist angles of the director a modulated phase emerges from the initial state above a definite threshold voltage. The periodic distortions are visible as a striped texture. Thus three phases have to be taken into consideration, namely the initial film state, the distorted state after a Freedericksz transition and a modulated phase. Allender has claimed [5] that these phases meet at a Lifshitz point in a suitably constructed phase diagram. Close to Lifshitz points the wavevector of the modulated phase tends to

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zero. Exactly the same behaviour has also been predicted for nematic films, when the ratio of the elastic constants for twist and splay distortions approaches a critical value [2, 6].

In this paper we discuss the main results of an amplitude equation for cholesteric film distortions, which is valid in the vicinity of Lifshitz points. More involved calculations deriving this equation are presented in [7]. It turns out that a criterion can be applied to decide, whether a striped texture appears after applying an electric field or not. Such a criterion is also of practical importance, as cholesteric films are widely used in display devices [8]. The occurrence of a periodic pattern can inhibit the application of strongly twisted cholesteric films [9].

Figure 1 shows, schematically, the geometry of a cholesteric layer which is confined between the bounding plates $X = 0$ and $X = d$. The tilt angle, θ, is enclosed between the preferred direction of the long molecular axes (the director) and the plane X = constant. At the lower and upper plate θ has fixed values η_1 and η_2, respectively. The azimuthal angle Φ grows gradually with increasing X by α.

Periodic distortions are oriented with the wavevector parallel to the Y axis of a cartesian coordinate systems, so that the stripes are parallel to the Z axis. The wavevector of the distortions and the projection of the director on to the lower plate enclose a definite angle $\mathscr{K}$. For convenience we shall use dimensionless coordinates

$$x = \frac{\pi X}{d} \quad \text{and} \quad y = \frac{\pi Y}{d}. \tag{1}$$

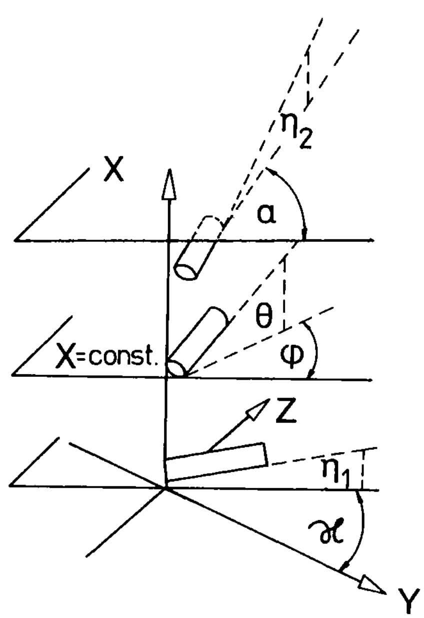

Figure 1. Geometry of a twisted cholesteric film. θ, is the angle between the director and the plane X = constant; Φ is the azimuthal angle of the director; α is the maximum twist angle; η_1, η_2, are the surface tilt angles; and $\mathscr{K}$, is the angle between the wavevector of the periodic distortions and the director projection on to the lower plate

2. Freedericksz transition

Near to the threshold voltage the Freedericksz transition of a cholesteric film can be described by an analytical theory [10]. We introduce the notation

$$\left.\begin{aligned} k_2 &= \frac{K_{22}}{K_{11}}, & k_3 &= \frac{K_{33}}{K_{11}}, \\ \omega &= \frac{\alpha}{\pi}, & \beta &= \frac{2\pi d}{P\alpha}, \\ \gamma &= \frac{\varepsilon_{\|} - \varepsilon_{\perp}}{\varepsilon_{\perp}} \text{ and } & U_0 &= \sqrt{\left(\frac{\pi^2 K_{11}}{\varepsilon_{\|} - \varepsilon_{\perp}}\right)}, \end{aligned}\right\} \tag{2}$$

where K_{11}, K_{22} and K_{33} are elastic constants defined in the framework of the Oseen–Frank theory [1], P is the helix pitch of the cholesteric material, $\varepsilon_{\|}$ and $\varepsilon_{\perp}$ are dielectric constants measured parallel and perpendicular to the director, respectively ($\varepsilon_{\|} > \varepsilon_{\perp}$). When the surface tilt angles η_1 and η_2 are zero, the Freedericksz threshold voltage is [11]

$$U_F = U_0\sqrt{R}, \tag{3}$$

with

$$R = 1 + \omega^2(k_3 - 2k_2 + 2k_2\beta). \tag{4}$$

The voltage ratio

$$\mu = \frac{U - U_F}{U_F} \tag{5}$$

is assumed to be small in all of the formulae presented in this paper. Here U is the effective value of an alternating voltage applied across the thin layer. Close to the Freedericksz threshold of the cholesteric film the director orientation is determined by

$$\left.\begin{aligned} \Theta &= b_0 \sin x, \\ \Phi &= \omega\left[x + \left(\frac{k_3 - 2k_2 + k_2\beta}{4k_2}\right) b_0^2 \sin 2x\right]. \end{aligned}\right\} \tag{6}$$

According to Raynes [11] the distortion amplitude b_0 obeys the equation $Bb_0^2 = \mu R$, where

$$B = \frac{1}{4}\left\{k_3 + R\gamma - \omega^2\left[\frac{(k_3 - 2k_2 + k_2\beta)^2}{k_2} + 3(k_3 - k_2)\right]\right\}. \tag{7}$$

Depending on the sign of B either a supercritical ($B > 0$) or a subcritical bifurcation ($B < 0$) results. If there are small non-zero surface tilt angles η_1 and η_2, then b_0 satisfies the extended equation [10]

$$-\mu R b_0 + B b_0^3 = \frac{2\eta}{\pi}, \tag{8}$$

with

$$\eta = \tfrac{1}{2}(\eta_1 + \eta_2).$$

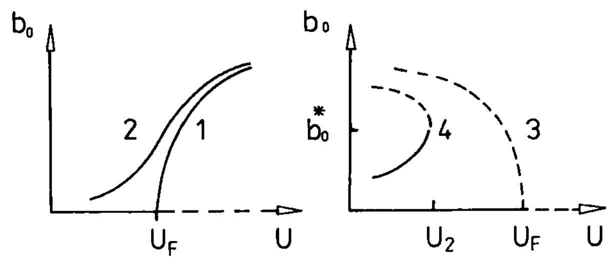

Figure 2. Schematic representation of bifurcation diagrams. ——, stable branches; - - -, unstable branches; (1) $B > 0, \eta = 0$; (2) $B > 0, \eta \neq 0$; (3) $B < 0, \eta = 0$; (4) $B < 0, \eta \neq 0$; b_0, is the distortion amplitude and U, is the applied voltage.

Plotting b_0 versus the applied voltage U we obtain bifurcation diagrams presented in figure 2. When $\eta \neq 0$ and $B > 0$ film distortions grow rapidly in a relative narrow voltage interval. In the opposite case $B < 0$ the film becomes unstable at a turning point ($U = U_2$), so that the director configuration changes discontinuously towards a strongly distorted state.

3. Amplitude equation for long wavelength modulations

A general mathematical description for the transition of cholesteric films to modulated equilibrium structures is rather complicated. However, simplifications are possible when the wavevector of the periodic distortions is small. As is well known from the theory of multicritical points [12, 13], long wavelength modulations appear in the vicinity of Lifshitz points. Actually, provided that $\eta = 0$, such points exist in the phase diagrams for cholesteric film instabilities [14].

The influence of weak boundary tilt angles ($|\eta| \ll 1$) on the phase diagrams is discussed separately in §6.

To lowest order of magnitude the perturbation theory [7] leads to

$$\left.\begin{aligned} \Theta &= b(y)\sin x, \\ \Phi &= \Omega + F(x)b_y, \end{aligned}\right\} \tag{9}$$

where

$$\Omega = \omega x + \mathscr{H}$$

and

$$b_y = \frac{db}{dy}.$$

(Now the origin of Φ is the y axis, so that Φ $(x = 0) = \mathscr{H}$ at the lower plate and Φ $(x = \pi) = \mathscr{H} + \alpha$ at the upper one.) The distortion amplitude $b(y)$ obeys the differential equation

$$-Ab_{yyyy} + \delta b_{yy} + \mu R b - Bb^3 = 0, \tag{10}$$

which is valid for small control parameters δ and μ. $F(x)$ and δ are defined in the following way. Introducing an integral operator In by

$$\begin{aligned} \mathrm{In}(f(x)) = &- \frac{\pi - x}{\pi}\int_0^x \xi f(\xi)\, d\xi \\ &- \frac{x}{\pi}\int_x^\pi (\pi - \xi) f(\xi)\, d\xi, \end{aligned} \tag{11}$$

we find

$$F(x) = \ln(f(x)), \tag{12}$$

where

$$f(x) = \left(\frac{1-k_2}{k_2}\right)\sin\Omega\cos x - \left(\frac{k_3 - k_2 + 2k_2\beta}{k_2}\right)\omega\cos\Omega\sin x. \tag{13}$$

Analogously, we define

$$w(x) = \gamma\cos\Omega\sin x \tag{14}$$

and

$$W(x) = \ln(w(x)). \tag{15}$$

Using the expression

$$M(x) = (1-k_2)\sin\Omega\frac{dF(x)}{dx} + (1 + k_3 - 2k_2 + 2k_2\beta)\omega\cos\Omega F(x) - (k_2\sin^2\Omega + k_3\cos^2\Omega)\sin x + R\cos\Omega W(x), \tag{16}$$

a parameter dependent integral

$$\delta^*(\mathscr{H}) = -\frac{1}{\pi}\int_0^\pi dx\sin x M(x) \tag{17}$$

is obtained. Let $\delta^*(\mathscr{H})$ have its absolute minimum for $\mathscr{H} = \mathscr{H}_m$. If $\mathscr{H}_m$ obeys

$$\mathscr{H}_m = \begin{cases} 0 \text{ or } \dfrac{\pi}{2} & \text{if } \alpha = 0, \\ -\dfrac{\alpha}{2} + n\pi & \text{if } \alpha \neq 0, \end{cases} \quad (n \text{ is an integer}), \tag{18}$$

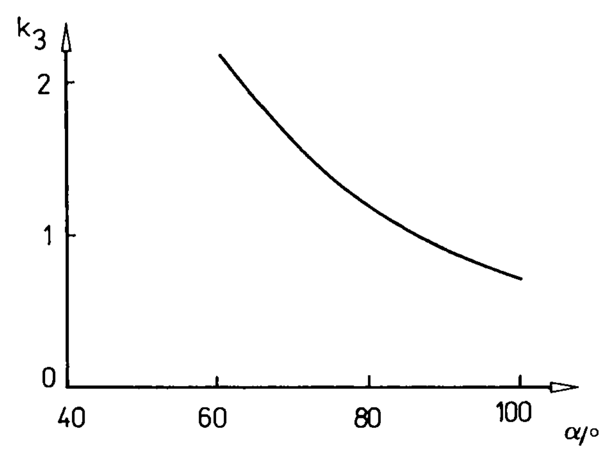

Figure 3. Equation (10) is applicable in the region above a curve in a $k_3 - \alpha$ diagram. Here such a limit is drawn for the case $\gamma = 1$ and $\beta = 0{\cdot}5$.

then the coefficient δ is determined by

$$\delta = \delta^*(\mathscr{H} = \mathscr{H}_m). \tag{19}$$

Equation (18) is a self-consistency condition for the perturbation theory derived in [7] and turns out to be valid in a large region of the parameter space. However, as we can see in figure 3 condition (18) is not satisfied for small non-zero angles α and small values of k_3. A procedure to determine the coefficient A in equation (10) is presented in the Appendix.

4. Phase diagram with a Lifshitz point

An investigation of equation (10) reveals that there is a competition between the Freedericksz transition with threshold $\mu_F = 0$ and a transition to a modulated structure, which appears at a threshold $\mu_S < 0$ when the condition

$$\delta < 0 \tag{20}$$

is satisfied. Then the uniform state $b = 0$ is unstable with respect to long wavelength modulations. Generally, inequality (20) is sufficient but not necessary for the existence of a modulated phase. However, if the conditions

$$A > 0 \quad \text{and} \quad B > 0 \tag{21}$$

are also satisfied, equation (10) describes the film states in the vicinity of a Lifschitz point [12, 13], whose position is determined by

$$\mu = 0 \quad \text{and} \quad \delta = 0. \tag{22}$$

Close to Lifshitz points a detailed description of different phases is possible in a simple manner. δ depends on several material parameters. It is convenient to choose k_2 as an independent variable. Let K be the critical value of k_2, which is obtained by $\delta(k_2 = K) = 0$. When terms proportional to $(k_2 - K)^2$ are neglected, we can write

$$\delta = a(k_2 - K), \tag{23}$$

where

$$a = \left(\frac{\partial \delta}{\partial k_2}\right)_K \tag{24}$$

is usually positive. Now the phase diagram shown in figure 4 results from equation (10) according to the general theory of Lifshitz points [13]. Obviously, if $k_2 > K$ only the Freedericksz transition is possible, whereas in the opposite case $k_2 < K$ a modulated phase emerges from the initial director configuration at a voltage below the Freedericksz threshold U_F. The modulated phase is stable within the voltage interval

$$\mu_1 < \frac{U}{U_F} - 1 < \mu_2, \tag{25}$$

where

$$\left.\begin{aligned} \mu_1 &= -\frac{\delta^2}{4AR}, \\ \mu_2 &= \frac{\delta^2}{\sqrt{2AR}}. \end{aligned}\right\} \tag{26}$$

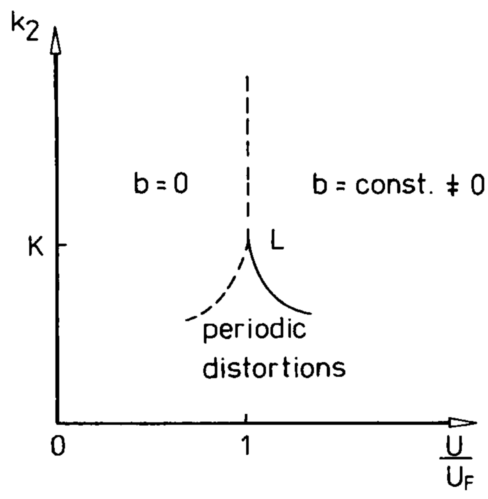

Figure 4. Phase diagram of a cholesteric layer containing a Lifshitz point L. ——, discontinuous transition; – – –, continuous transition.

5. Criterion for the occurrence of the modulated phase

The set of Lifshitz points obtained by the equation $\delta(k_2, k_3) = 0$ defines a line in a $k_2 - k_3$ diagram, when the other parameters α, β and γ are fixed. Above this line $k_2 > K$ is obeyed and according to figure 4 only the Freedericksz transition takes place. Below the line of Lifshitz points a periodic pattern appears. Thus there is a simple possibility to decide, whether a striped texture emerges from the initial state $b = 0$ or not.

As illustrated in figures 5, 6 and 7 the existence region of the modulated phase becomes more extended by increasing α and β and reducing γ. All of the curves plotted in these figures are sets of Lifshitz points obeying conditions (21). In other cases, when the conditions (21) are not satisfied, criterion (20) should be also useful to estimate the existence region of a modulated structure in the film.

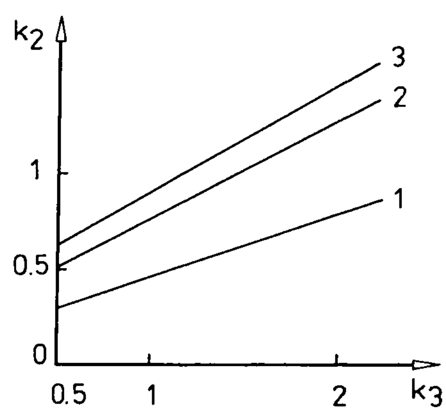

Figure 5. Lines of Lifshitz points for different twist angles. Below each line in the diagram a modulated phase is stable within the voltage interval (equation (25)). In the region above a line only the Freedericksz transition takes place. $\beta = 0{\cdot}67$ and $\gamma = 1$; (1) $\alpha = 135°$; (2) $\alpha = 200°$; (3) $\alpha = 225°$.

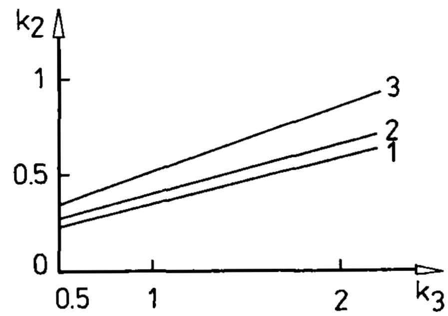

Figure 6. Lines of Lifshitz points for $\alpha = 135°$ and $\gamma = 1$, (1) $\beta = 0{\cdot}33$; (2) $\beta = 0{\cdot}5$; (3) $\beta = 0{\cdot}75$.

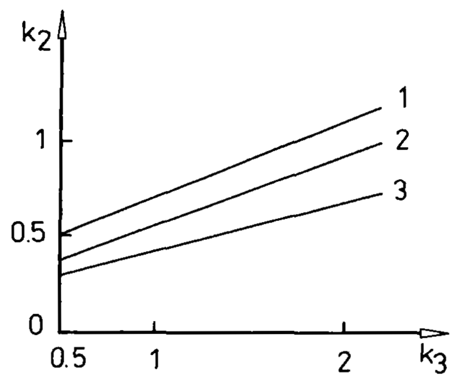

Figure 7. Lines of Lifshitz points for $\alpha = 180°$ and $\beta = 0{\cdot}5$. (1) $\gamma = 0{\cdot}5$; (2) $\gamma = 1{\cdot}0$; (3) $\gamma = 2{\cdot}0$.

6. Small pretilt angles at the boundaries

More realistic boundary conditions imply small surface tilt angles η_1 and η_2 (see figure 1). For this case an extended time dependent amplitude equation

$$b_\tau + Ab_{yyyy} - \delta b_{yy} - \mu Rb + Bb^3 - \frac{2\eta}{\pi} = 0 \tag{27}$$

can be derived [7], where τ is the time measured in dimensionless units. Because of the symmetry breaking term $2\eta/\pi$ continuous phase transitions do not occur and any film instabilities are accompanied with a jump of b. Provided that $A > 0$, equation (27) is suitable to determine the condition for neutral stability of the uniform state $b = b_0$. Inserting in equation (27) the transformation

$$b(\tau, y) = b_0 + s(\tau, y) \tag{28}$$

and taking into account equation (8) we find

$$s_\tau = -As_{yyyy} + \delta s_{yy} + (\mu R - 3b_0^2 B)s, \tag{29}$$

where non-linear terms are omitted.

Stability of the uniform state implies that any possible perturbation with wavenumber q

$$s = s_0 \exp(p\tau + iqy) \tag{30}$$

relaxes back ($p(q) < 0$). If the initial state $b = b_0$ is unstable, the wavenumber for the fastest growing mode is

$$q_m = \sqrt{-\frac{\delta}{2A}} \ll 1. \tag{31}$$

Neutral stability is achieved when

$$p(q_m) = 0. \tag{32}$$

As $A > 0$ we conclude from equation (31) that condition $\delta < 0$ is necessary for the occurrence of a periodic pattern. Combining equations (29), (30), (31) and (32) yields

$$\delta^2 = 4A(3b_0^2B - \mu R)$$

and eliminating μR by using equation (8) we find

$$\delta^2 = 8A\left(\frac{\eta}{\pi b_0} + Bb_0^2\right). \tag{33}$$

Equations (8) and (33) represent the condition of neutral stability for the homogeneous film state $b = b_0$. However, it should be noted, that we only consider the case of small surface tilt angles obeying

$$\eta^{1/3} \ll (2\pi|B|)^{1/3}. \tag{34}$$

6.1. *Line of neutral stability for $B > 0$*

Using $b_0 > 0$ as a freely varying parameter in equations (8) and (33), a line of neutral stability in a plot of k_2 versus $U/U_F = 1 + \mu$ can be computed. This line encloses the region in which the film state $b = b_0$ is unstable and a striped texture must occur. The diagram in figure 8 refers to a nematic film with $\alpha = 0$. However the topological features of the diagrams are not changed if $\alpha \neq 0$ as long as conditions (21) are satisfied. It can be seen by comparing figures 8 and 4, that the phase diagrams for $\eta \neq 0$ differ considerably from those for $\eta = 0$, as in the former case a Freedericksz transition with a sharp threshold does not occur.

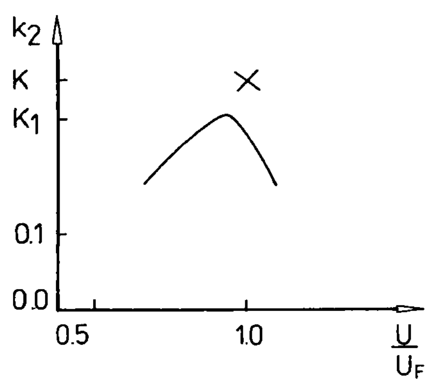

Figure 8. Phase diagram for non-zero surface tilt angles ($\eta \neq 0$) and $B > 0$. Below the lines of neutral stability a modulated phase must appear. Here this line is drawn for the case $\alpha = 0, k_3 = 1{\cdot}5, \gamma = 1$ and $\eta = 0{\cdot}1°$. The cross indicates the position of a Lifshitz point appearing when $\eta = 0$.

Of practical interest is the maximum value K_1 of k_2 at which the phase region of the striped texture ends (see figure 8). A straightforward calculation leads to the result

$$K_1 = K - L\eta^{1/3}, \tag{35}$$

where

$$L = \left(\frac{12^{1/2}2^{1/6}}{\pi^{1/3}}\right)\frac{A^{1/2}B^{1/6}}{a} \tag{36}$$

and K is obtained from the equation $\delta(k_2 = K) = 0$. Obviously, small surface tilt angles shift considerably the phase region of periodic distortions toward lower values of k_2. It is also possible to determine a critical value

$$\eta_c = \frac{\pi}{24\sqrt{6}}\frac{|\delta^3|}{\sqrt{(A^3B)}} \tag{37}$$

of η above which periodic distortions are completely suppressed. For example, if $k_2 = 0{\cdot}5$, $k_3 = 1{\cdot}5$, $\alpha = 180°$, $\beta = 0{\cdot}6$ and $\gamma = 1{\cdot}5$ film modulations disappear for $\eta > \eta_c$ with $\eta_c = 0{\cdot}5°$.

6.2. *Case $B < 0$*

If $B < 0$, a modulated phase always exists for $\delta < 0$ (or $k_2 < K$) independently of η. (However this conclusion is restricted to small surface tilt angles obeying condition (34).) Then periodic distortions should be stable within the voltage interval

$$1 + \mu_1 \leqslant \frac{U}{U_F} \leqslant 1 + \mu_2, \tag{38}$$

where μ_1 and μ_2 are defined by

$$\mu_i = -\frac{1}{R}\left(\frac{2\eta}{\pi b_i} - Bb_i^2\right), \quad (i = 1, 2) \tag{39}$$

with

$$\left.\begin{aligned} b_1 &= \left(-\frac{\eta}{2\pi B} + \sqrt{D}\right)^{1/3} - \left(\frac{\eta}{2\pi B} + \sqrt{D}\right)^{1/3}, \\ b_2 &= \left(-\frac{\eta}{\pi B}\right)^{1/3} \end{aligned}\right\} \tag{40}$$

and

$$D = \left(-\frac{\delta^2}{24AB}\right)^3 + \left(\frac{\eta}{2\pi B}\right)^2.$$

$b_2 \equiv b_0^*$ is the value of b_0 at the turning point of curve (4) in figure 2.

The voltage interval (38) is often rather small. In the special case $\eta = 0$ we find

$$1 - \frac{\delta^2}{4AR} \leqslant \frac{U}{U_F} \leqslant 1. \tag{41}$$

For example, if $\alpha = 180°$, $\eta = 0$, $k_2 = 0{\cdot}5$, $k_3 = 1{\cdot}5$, $\beta = 0{\cdot}6$ and $\gamma = 1$ periodic distortions occur in the narrow region $0{\cdot}97 \leqslant U/U_F \leqslant 1$. This region becomes still smaller for non-zero surface tilt angles.

With increasing twist angle α beyond 180° the value of A is reduced and the phase region of the modulated structure is extended considerably. However if A is too small, or if $A < 0$, the analytic theory fails, as the wavenumber q_m for the fastest growing distortion mode is no longer defined by relation (31).

7. Comparison with experimental results

Chigrinov *et al.* [4] have investigated, both experimentally and theoretically, cholesteric films in wedge-shaped cells. Since the film thickness varies in the cell, there is also a variation of β in each Grandjean zone, whereas the other parameters $k_2 = 0{\cdot}67$, $k_3 = 1{\cdot}25$ and $\gamma \approx 0$ are fixed ($\eta = 0$). The experimental results confirm, that equation (18) for the angle $\mathscr{H}_m$ is valid. If $\alpha = 180°$ the parameter β must satisfy $0{\cdot}5 \leqslant \beta \leqslant 1{\cdot}5$. For this Grandjean zone condition (20) is obeyed and a modulated phase results in agreement with the findings in [4]. It turns out that the Lifshitz point is not observable in the present case, because for the critical value $\beta_L = 0{\cdot}2$ a film with twist angle $\alpha = 180°$ is not stable.

As a second example we consider the case $\alpha = 90°$ and $0 \leqslant \beta \leqslant 2$. From condition $\delta = 0$ we find the critical value $\beta_L = 0{\cdot}84$ for the Lifshitz point. Actually, figure 6 in [4] suggests that the wavenumber q_m goes to zero in the Grandjean zone with twist angle $\alpha = 90°$ for the wedge-shaped cell. However more sensitive experiments for checking the theory would be desirable.

Appendix

Coefficient A in differential equation (10)

The functions $F(x)$, $W(x)$ and $M(x)$ are already defined in the text. Furthermore, introducing

$$P(x) = M(x) - \frac{2}{\pi}\sin x \int_0^{\pi} d\xi \sin \xi M(\xi), \tag{A 1}$$

we obtain

$$D(x) = \int_0^{x} d\xi \sin (x - \xi) P(\xi). \tag{A 2}$$

In the next step the functions $C(x)$ and $H(x)$ are defined by

$$C(x) = \ln (c(x)), \tag{A 3}$$

where

$$c(x) = \gamma D(x) \cos \Omega - (1 + \gamma \cos^2 \Omega) W(x) \tag{A 4}$$

and

$$H(x) = \ln (h(x)), \tag{A 5}$$

where

$$k_2 h(x) = -(\sin^2 \Omega + k_3 \cos^2 \Omega) F(x) + (1 - k_2) \sin \Omega \frac{dD(x)}{dx} - (k_3 - k_2 + 2k_2\beta)\omega \cos \Omega D(x). \tag{A 6}$$

Finally, A is obtained by integration

$$A = \frac{1}{\pi}\int_0^{\pi} dx \sin x T(x), \tag{A 7}$$

with

$$T(x) = \omega \cos \Omega (1 + k_3 - 2k_2 + 2k_2\beta) H(x) + (1 - k_2) \sin \Omega \frac{dH(x)}{dx}$$
$$- (k_2 \sin^2 \Omega + k_3 \cos^2 \Omega) D(x) + R \cos \Omega C(x). \quad \text{(A 8)}$$

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