On the Electric Field Induced Stripe Instability Threshold of Twisted Nematic Layers

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Based on continuum theory, general expressions are derived which indicate how the electric field induced stripe instability threshold of twisted nematic layers is influenced by the liquid crystal material parameters. Results are applied to supertwist displays.

1. Introduction

Most liquid crystal displays make use of the electrooptic properties of twisted nematic layers. Both the standard twisted nematic displays of 90° twist [1] as well as the recently developed 'supertwist' displays of more than 180° twist [2] require director fields which depend only on one spatial coordinate, the coordinate z parallel to the layer normal. Director fields which depend on more than one spatial coordinate may, however, also be stable in twisted layers. They appear as periodic structures, such as parallel stripes [3], which make them unsuitable for display applications. Recent experiments on polymer liquid crystals in magnetic fields have shown that stripe formation may not be restricted to layers of relatively high twist, but can even occur when the twist is zero [4]. For stripes to form with zero twist, the twist to splay elastic constant ratio k_{22}/k_{11} must be smaller than the critical value [4–8]

$$\frac{k_{22}}{k_{11}} < 1 - \frac{(1 - 8/\pi^2)^{1/2}}{1 + (1 - 8/\pi^2)^{1/2}} \approx 0.303. \qquad (1)$$

Even though analogous experiments in electric fields have not yet been done, theory predicts that (1) is also valid for electric fields [7]. For zero twist the direction of the stripes was found to be parallel to the director orientation at the layer surfaces.

For layers of nonzero twist, the criterion for the occurrence of stripes is much more complicated. Numerical as well as experimental results indicate that, in

addition to k_{22}/k_{11}, the values of the bend to splay elastic constant ratio k_{33}/k_{11} and the relative dielectric anisotropy $\gamma = \Delta\varepsilon/\varepsilon_\perp$ are important for stripe formation [3, 9–11]. For 180° twisted layers, stripe domains occur when the following inequality holds [12, 13]:

$$4\left(\frac{k_{22}}{k_{11}}\right)^2 + 12\frac{k_{22}}{k_{11}}\frac{k_{33}}{k_{11}} + 8\left(1 - \frac{k_{22}}{k_{11}}\right)^2$$
$$- 3\left(1 - \frac{k_{33}}{k_{11}} - 4\frac{k_{22}}{k_{11}}\frac{d}{p}\right)^2 - \frac{2}{3}(3 + \pi^2)u_F^4$$
$$+ \frac{1}{3}(15 + 2\pi^2)\frac{k_{22}}{k_{11}}\gamma u_F^2 < 0. \qquad (2)$$

Here d is the layer thickness, p the natural pitch (for 2π rotation) and u_F the reduced Fréedericksz threshold given by

$$u_F = \left(1 + \frac{k_{33}}{k_{11}} - 2\frac{k_{22}}{k_{11}} + 4\frac{k_{22}}{k_{11}}\frac{d}{p}\right)^{1/2}. \qquad (3)$$

The stripe orientation was found to be perpendicular to the midplane director of the undisturbed layer (i.e. zero voltage). In this study, we will generalize (2) for the case of arbitrary twist angles ϕ_T and show how the various parameters influence the tendency for stripe formation in supertwist displays.

2. Basic Equations

Consider a twisted layer of thickness d of a chiral nematic ('cholesteric') material of pitch p with a positive dielectric anisotropy $\Delta\varepsilon$. The layer is confined between the planes $z = \pm d/2$ of a Cartesian coordinate system (x, y, z). The optic axis orientation of the

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liquid crystal is described by the director field

$$L = (\cos\theta\cos\phi,\ \cos\theta\sin\phi,\ \sin\theta), \tag{4}$$

where, in general, the tilt angle θ and the twist angle ϕ are functions of x, y, and z. If no external field is applied, the layer is assumed to have a uniformly twisted structure with

$$\theta = 0, \qquad \phi = \phi_T \frac{z}{d} - \alpha. \tag{5}$$

ϕ_T is the total twist and α is constant. The orientation at the boundaries $z = \pm d/2$ is given by

$$
\begin{aligned}
&L\,(x,\,y,\,\pm d/2) \\
&= (\cos(\pm\phi_T/2-\alpha),\ \sin(\pm\phi_T/2-\alpha),\ 0).
\end{aligned} \tag{6}
$$

In the middle of the layer $(z=0)$, the director is

$$L_m = (\cos\alpha,\ -\sin\alpha,\ 0), \tag{7}$$

making an angle of $-\alpha$ with respect to the x axis. In the case of stripe formation we choose the direction of the x axis parallel to the stripes, and α is thus the angle of the stripe direction relative to the midplane director L_m of the uniformly twisted structure. With this choice of the coordinates, the director field depends only on y and z. If a voltage U is applied across the layer, it follows that the electric field can be expressed as

$$E = (0,\ E_y,\ E_z). \tag{8}$$

The components E_y and E_z, which are functions of y and z, are related to each other and to the applied voltage by the three equations

$$\frac{\partial E_y}{\partial z} = \frac{\partial E_z}{\partial y}, \quad \int_{-d/2}^{d/2} E_z\,dz = U, \quad \operatorname{div}(\hat\varepsilon\, E) = 0, \tag{9}$$

where the dielectric tensor $\hat\varepsilon$ is a function of L. The third equation of (9) implies that the electric conductivity of the liquid crystal is negligible. To simplify the mathematical expressions, we introduce dimensionless coordinates and voltages:

$$\eta = y/d, \quad \zeta = z/d, \quad u = U/U_{11}, \tag{10}$$

where $U_{11} = \pi\left(\dfrac{k_{11}}{\varepsilon_0\,\Delta\varepsilon}\right)^{1/2}$ is the well known Fréedericksz threshold for nontwisted layers.

As shown in [3], the threshold voltage u_s for stripe formation can be obtained from a small angle expansion of the torque equation governing the director field. For a periodic strip pattern of period Λ, we use the following small angle approximations for the angles θ and ϕ and the electric field components:

$$
\begin{aligned}
\theta(\eta,\zeta) &= \delta\sin(2\pi q\eta)\,W(\zeta), \\
\phi(\eta,\zeta) &= \phi_T\zeta - \alpha + \delta\cos(2\pi q\eta)\,V(\zeta), \\
E_\eta(\eta,\zeta) &= -\delta\,(U/d)\,2\pi q\sin(2\pi q\eta)\,F(\zeta), \\
E_\zeta(\eta,\zeta) &= (U/d)\,[1 + \delta\cos(2\pi q\eta)\,F'(\zeta)],
\end{aligned} \tag{11}
$$

where $\delta \ll 1$, $q = d/\Lambda$ and $F' = dF/d\zeta$. The last two equations of (11) are compatible with the first equation of (9). The case $\delta = 0$ corresponds to the uniformly twisted structure. In lowest order of δ, (11) leads to the following homogeneous system of coupled differential equations for the unknown functions W, V, and F:

$$
\begin{aligned}
&-\frac{W''}{\pi^2} + (u_F^2 - 1 - u^2)\,W + 2q^2\left[\left(\frac{k_{33}}{k_{11}} + \frac{k_{22}}{k_{11}}\right)\right.\\
&\qquad \left. - \left(\frac{k_{33}}{k_{11}} - \frac{k_{22}}{k_{11}}\right)\cos 2(\phi_T\zeta-\alpha)\right]W \\
&- 2q\left[\left\{\left(1 + \frac{k_{33}}{k_{11}} - 2\frac{k_{22}}{k_{11}}\right)\frac{\phi_T}{\pi}\right.\right.\\
&\qquad \left.+ 4\frac{k_{22}}{k_{11}}\frac{d}{p}\right\}\sin(\phi_T\zeta-\alpha)\,V \\
&\qquad - \left(1 - \frac{k_{22}}{k_{11}}\right)\cos(\phi_T\zeta-\alpha)\frac{V'}{\pi} \\
&\qquad \left. - \pi u^2\sin(\phi_T\zeta-\alpha)\,F\right] = 0, \\[6pt]
&-\frac{V''}{\pi^2} + 2q^2\left[\left(\frac{k_{33}}{k_{11}} + 1\right)\right.\\
&\qquad \left. - \left(\frac{k_{33}}{k_{11}} - 1\right)\cos 2(\phi_T\zeta-\alpha)\right]\left(\frac{k_{22}}{k_{11}}\right)^{-1}V \\
&- 2q\left[\left\{\left(\frac{k_{33}}{k_{11}} - \frac{k_{22}}{k_{11}}\right)\frac{\phi_T}{\pi}\right.\right.\\
&\qquad \left.+ 4\frac{k_{22}}{k_{11}}\frac{d}{p}\right\}\sin(\phi_T\zeta-\alpha)\,W \\
&\qquad \left. + \left(1 - \frac{k_{22}}{k_{11}}\right)\cos(\phi_T\zeta-\alpha)\frac{W'}{\pi}\right]\left(\frac{k_{22}}{k_{11}}\right)^{-1} = 0, \\[6pt]
&-\frac{F''}{\pi^2} + 2q^2\,[(2+\gamma) - \gamma\cos 2(\phi_T\zeta-\alpha)]\,F \\
&\qquad - 2q\frac{1}{\pi}\gamma\sin(\phi_T\zeta-\alpha)\,W = 0.
\end{aligned} \tag{12}
$$

Here u_F denotes the reduced Fréedericksz threshold for arbitrary twist angles ϕ_T: (13)

$$u_F = \left[1 + \left(\frac{k_{33}}{k_{11}} - 2\frac{k_{22}}{k_{11}}\right)\left(\frac{\phi_T}{\pi}\right)^2 + 4\frac{k_{22}}{k_{11}}\frac{d}{p}\frac{\phi_T}{\pi}\right]^{1/2}.$$

The first two equations of (12) follow from standard Oseen-Frank theory [14] and the third equation of (12) is obtained from the third equation of (9). The system (12) has to be solved subject to the boundary conditions

$$W(\pm 1/2) = V(\pm 1/2) = F(\pm 1/2) = 0. \quad (14)$$

For a given q, solutions can only be found for discrete values (eigenvalues) of the reduced voltage u. In order for solutions to be stable it is required that $du/dq = 0$ [3].

3. Expansion for Small Wave Vectors ($q \ll 1$)

In order to determine under which conditions stripe formation may occur at voltages lower than the Fréedericksz threshold, we solve (12) for $q \ll 1$. Using the expansions

$$W(\zeta) = \cos(\pi\zeta) + q^2 W_2(\zeta),$$
$$V(\zeta) = q V_1(\zeta),$$
$$F(\zeta) = q F_1(\zeta), \quad (15)$$
$$u^2 = u_F^2 + q^2 \Gamma,$$

the second and third equations of (12) may be solved for V_1 and F_1 by straightforward integration. Expansion of W_2 into a Fourier series (with respect to ζ) which is compatible with (14), subsequent multiplication of the first equation of (12) with $\cos(\pi\zeta)$ and integration over ζ from $-1/2$ to $1/2$ leads to

$$\Gamma = \Gamma_1 \cos^2\alpha + \Gamma_2 \sin^2\alpha \quad (16)$$

with the Γ_i being functions of ϕ_T, d/p, k_{33}/k_{11}, k_{22}/k_{11}, and γ. We distinguish the following three cases:

a) $\Gamma_1 > 0$, $\Gamma_2 > 0$,

b) $\Gamma_1 < 0$, $\Gamma_1 < \Gamma_2$, $\quad (17)$

c) $\Gamma_2 < 0$, $\Gamma_2 < \Gamma_1$.

In the first case, the stripe threshold is higher than the Fréedericksz threshold, i.e. no stripes occur. In the last two cases, the stripe threshold is lower, with the stripes being parallel to the midplane director (see above) for b) and perpendicular for c). The following equation has been derived for the Γ_i:

$$\Gamma_i = 2\left(\frac{k_{33}}{k_{11}} + \frac{k_{22}}{k_{11}}\right) - 2\left(\frac{k_{33}}{k_{11}} - \frac{k_{22}}{k_{11}}\right) J_0^{(i)}$$
$$+ \left[\left(1 + \frac{k_{33}}{k_{11}} - 2\frac{k_{22}}{k_{11}}\right)\frac{\phi_T}{\pi} + 4\frac{k_{22}}{k_{11}}\frac{d}{p}\right]\left(\frac{k_{22}}{k_{11}}\right)^{-1}$$

$$\cdot\left[\left\{1 - \frac{k_{22}}{k_{11}} + \left(\frac{k_{33}}{k_{11}} - \frac{k_{22}}{k_{11}}\right)\frac{\phi_T}{\pi} + 4\frac{k_{22}}{k_{11}}\frac{d}{p}\right\} J_1^{(i)}\right.$$
$$\left. + \left\{1 - \frac{k_{22}}{k_{11}} - \left(\frac{k_{33}}{k_{11}} - \frac{k_{22}}{k_{11}}\right)\frac{\phi_T}{\pi} - 4\frac{k_{22}}{k_{11}}\frac{d}{p}\right\} J_2^{(i)}\right]$$
$$- \left(1 - \frac{k_{22}}{k_{11}}\right)\left(\frac{k_{22}}{k_{11}}\right)^{-1} \quad (18)$$

$$\cdot\left[\left\{1 - \frac{k_{22}}{k_{11}} + \left(\frac{k_{33}}{k_{11}} - \frac{k_{22}}{k_{11}}\right)\frac{\phi_T}{\pi} + 4\frac{k_{22}}{k_{11}}\frac{d}{p}\right\} J_3^{(i)}\right.$$
$$\left. + \left\{1 - \frac{k_{22}}{k_{11}} - \left(\frac{k_{33}}{k_{11}} - \frac{k_{22}}{k_{11}}\right)\frac{\phi_T}{\pi} - 4\frac{k_{22}}{k_{11}}\frac{d}{p}\right\} J_4^{(i)}\right]$$

$$+ \gamma u_F^2 (J_2^{(i)} - J_1^{(i)}),$$

where the $J_j^{(i)}$ depend only on the total twist ϕ_T:

$$J_0^{(1)} = -J_0^{(2)} = \frac{\pi^2 \sin\phi_T}{\phi_T(\pi^2 - \phi_T^2)},$$

$$J_1^{(1)} = \frac{-\pi^2}{(\pi - \phi_T)^2}\left[1 - \frac{\pi(\pi - 3\phi_T)}{\phi_T(\pi^2 - \phi_T^2)}\sin\phi_T\right.$$
$$\left. - \frac{32\pi\phi_T}{(\pi^2 - \phi_T^2)^2}\cos^2(\phi_T/2)\right],$$

$$J_2^{(1)} = \frac{\pi^2}{(\pi + \phi_T)^2}\left[1 - \frac{\pi(\pi + 3\phi_T)}{\phi_T(\pi^2 - \phi_T^2)}\sin\phi_T\right.$$
$$\left. + \frac{32\pi\phi_T}{(\pi^2 - \phi_T^2)^2}\cos^2(\phi_T/2)\right],$$

$$J_3^{(1)} = \frac{\pi}{(\pi - \phi_T)}\left[1 + \frac{\pi}{\phi_T(\pi - \phi_T)}\sin\phi_T\right.$$
$$\left. - \frac{16\pi}{(\pi^2 - \phi_T^2)(\pi - \phi_T)}\cos^2(\phi_T/2)\right],$$

$$J_4^{(1)} = \frac{\pi}{(\pi + \phi_T)}\left[1 + \frac{\pi}{\phi_T(\pi + \phi_T)}\sin\phi_T\right.$$
$$\left. - \frac{16\pi}{(\pi^2 - \phi_T^2)(\pi + \phi_T)}\cos^2(\phi_T/2)\right],$$

$$J_1^{(2)} = \frac{-\pi^2}{(\pi - \phi_T)^2}\left[1 + \frac{\pi(\pi - 3\phi_T)}{\phi_T(\pi^2 - \phi_T^2)}\sin\phi_T\right],$$

$$J_2^{(2)} = \frac{\pi^2}{(\pi + \phi_T)^2}\left[1 + \frac{\pi(\pi + 3\phi_T)}{\phi_T(\pi^2 - \phi_T^2)}\sin\phi_T\right], \quad (19)$$

$$J_3^{(2)} = \frac{\pi}{(\pi - \phi_T)}\left[1 - \frac{\pi}{\phi_T(\pi - \phi_T)}\sin\phi_T\right],$$

$$J_4^{(2)} = \frac{\pi}{(\pi + \phi_T)}\left[1 - \frac{\pi}{\phi_T(\pi + \phi_T)}\sin\phi_T\right].$$

The results given in (18) and (19) also apply to the case of magnetic fields (parallel to the layer normal), provided that u and u_F are replaced by the respective magnetic quantities in the fourth equation of (15) and that the last term is omitted in (18) (i.e. γ is set equal to zero). Concerning this last term, which is specific for electric fields, it can be shown that $(J_2^{(i)} - J_1^{(i)}) > 0$ for $\phi_T \neq 0$. It follows that the tendency for stripe formation generally decreases with increasing values of γ and that it is lower for electric fields than for magnetic fields. (Exceptions are stripe patterns with $\alpha = 0$ in non-twisted layers in which case $J_2^{(1)} - J_1^{(1)} = 0$). It can also be seen from (19) that $J_j^{(1)} = J_j^{(2)}$ whenever $\phi_T = \pm 3\pi$, $\pm 5\pi$, etc. This means that within the q^2 approximation of the fourth equation of (15), stripes with $\alpha = 0$ and $\pi/2$ have the same threshold. As was suggested earlier [15], this may explain the 'grid pattern' formation observed in 540° layers.

4. Special Cases: 0° and 180° Layers

It is seen from (19) that special attention is required when $\phi_T = 0$ or π. In the former case, the functions $J_j^{(i)}$ assume the values

$$J_0^{(1)} = 1, \quad J_1^{(1)} = J_2^{(1)} = 0, \quad J_3^{(1)} = J_4^{(1)} = 2 - (4/\pi)^2,$$

$$J_0^{(2)} = -1, \quad J_1^{(2)} = -J_2^{(2)} = -2, \quad J_3^{(2)} = J_4^{(2)} = 0. \quad (20)$$

For stripes to form in the director plane (i.e. $\alpha = 0$), the condition given by (1) is obtained from $\Gamma_1 < 0$. As mentioned, (1) is valid for electric as well as magnetic fields. For stripes perpendicular to the director plane ($\alpha = \pi/2$), we derive from $\Gamma_2 < 0$ the necessary condition

$$\frac{k_{33}}{k_{11}} < \left(4 \frac{k_{22}}{k_{11}} \frac{d}{p}\right)^2 - \gamma. \quad (21)$$

This inequality cannot be met by pure nematics ($d/p = 0$) since a negative elastic constant ratio would be required. For chiral nematics ($d/p \neq 0$), 'perpendicular stripes' are not forbidden by theory. Considering, however, that d/p is limited to 0.25 for $\phi_T = 0$, even for chiral nematics no perpendicular stripes can be expected since an unrealistically small ratio k_{33}/k_{11} would be required.

In the case $\phi_T = \pi$, the following values are derived for the quantities $J_j^{(i)}$:

$$J_0^{(1)} = 1/2, \quad J_1^{(1)} = 0, \quad J_2^{(1)} = 1/4, \quad J_3^{(1)} = 0, \quad J_4^{(1)} = 1/2,$$

$$J_0^{(2)} = -1/2, \quad J_1^{(2)} = -(1 + \pi^2/3)/2, \quad J_2^{(2)} = 3/4,$$

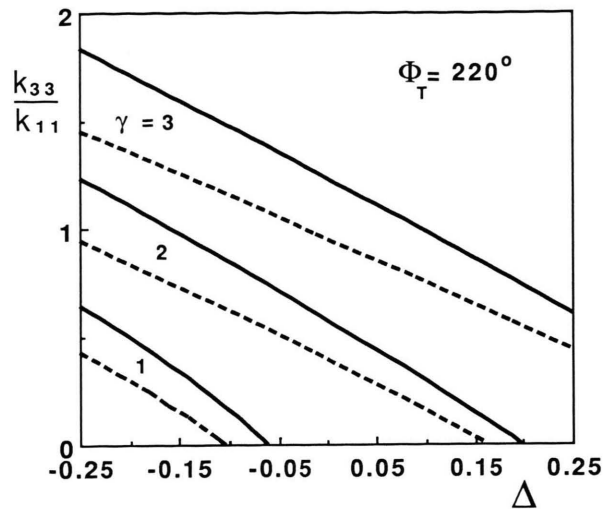$$J_3^{(2)} = -1, \quad J_4^{(2)} = 1/2. \quad (22)$$

Fig. 1. $\Gamma_2 = 0$ curves in the (Δ, k_{33}/k_{11}) plane for 220° twisted layers. Parameters are $\gamma = 1$, 2, and 3 and $k_{22}/k_{11} = 0.5$ (dashed lines) and 0.6 (solid lines).

From $\Gamma_1 < 0$ we find for stripes parallel to the midplane director

$$12 \left(\frac{k_{22}}{k_{11}}\right)^2 + 4 \frac{k_{22}}{k_{11}} \frac{k_{33}}{k_{11}} - \left(1 - \frac{k_{33}}{k_{11}} - 4 \frac{k_{22}}{k_{11}} \frac{d}{p}\right)^2$$

$$+ \frac{k_{22}}{k_{11}} \gamma u_F^2 < 0. \quad (23)$$

For perpendicular stripes, the condition (2) is obtained.

5. Application to Supertwist Displays

Numerical evaluation of (18) shows that, for material parameters and twist angles which are typical for supertwist displays, stripes only occur perpendicular to the midplane director ($\alpha = \pi/2$). For given twist angles ϕ_T, we have investigated the function Γ_2 (ϕ_T, Δ, k_{33}/k_{11}, k_{22}/k_{11}, γ), where Δ is defined as

$$\Delta = d/p - \phi_T/2\pi. \quad (24)$$

The parameter space was chosen as follows:

$$\pi \leq \phi_T \leq 3\pi/2, \quad -0.25 \leq \Delta \leq 0.25,$$

$$0 \leq k_{33}/k_{11} \leq 3, \quad 0.5 \leq k_{22}/k_{11} \leq 0.6, \quad 1 \leq \gamma \leq 3. \quad (25)$$

Typical results are presented in Figs. 1 and 2 for twist angles of 220° and 270°, respectively. Curves for $\Gamma_2 = 0$ are drawn in the (Δ, k_{33}/k_{11}) plane for γ values of 1, 2
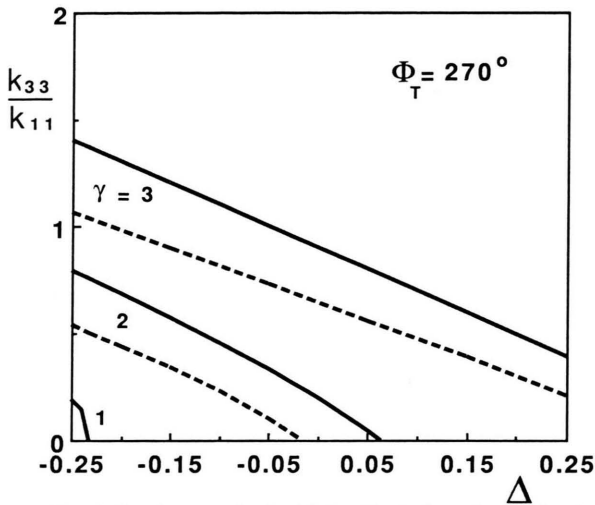
Fig. 2. $\Gamma_2 = 0$ curves in the $(\Delta, k_{33}/k_{11})$ plane for 270° twisted layers. Parameters as in Figure 1.
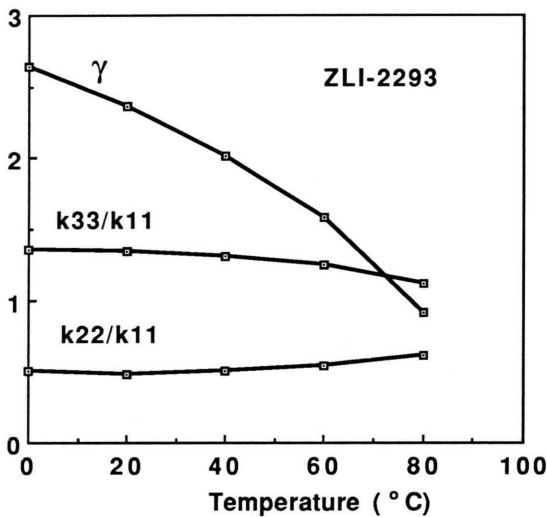

Fig. 3. Temperature dependence of the relative dielectric anisotropy γ and two elastic constant ratios of the liquid crystal mixture ZLI-2293 of Merck.

states with $\Gamma_2 > 0$ (no stripes). Let us now consider some point just above the curve. For the parameter space (25) it is found that decreasing Δ or k_{33}/k_{11} always has the effect of moving the point below the curve, i.e. into the region where the Fréedericksz configuration is stable. The same effect can be achieved by moving the curve upwards while keeping the point fixed in the $(\Delta, k_{33}/k_{11})$ plane. This implies increasing k_{22}/k_{11} or γ or decreasing ϕ_T. In agreement with the literature [9–11], we thus have the result that for supertwist displays the tendency for stripe formation may be reduced by decreasing ϕ_T, Δ or k_{33}/k_{11} and increasing k_{22}/k_{11} or γ. Since for all practical purposes the parameter space (25) seems to be sufficiently large, this result may be considered to be general. It has been stated before that changing parameters in order to prevent stripe domains in supertwist generally reduces the steepness of the contrast versus voltage characteristic [9–11]. This does not seem to be the case for the decrease of Δ.

In supertwist layers, stripe domain formation is sometimes observed at higher temperatures. In Fig. 3 the temperature dependence of three material parameters is shown for ZLI-2293 of Merck, a typical liquid crystal material for supertwist display application. It is noticed that the temperature change of the two elastic constant ratios is relatively small and in both cases in favor of lowering the stripe formation tendency. The temperature dependence of d/p (i.e. Δ) is also rather small [16, 17]. Assuming that temperature effects of the surface tilt angle can be neglected, it can be concluded from Fig. 3 that the observed stripe formation at higher temperatures is mainly caused by the strong temperature dependence of γ.

6. Further Work

Our analysis is not complete in that it is based on the assumption that the tilt angle of the twisted layer is zero on both boundaries. It would be of interest to incorporate nonzero pretilt angles in a theoretical treatment of the stripe formation threshold.

and 3, and k_{22}/k_{11} values of 0.5 (dashed curves) and 0.6 (solid curves). Consider a curve in Figs. 1 or 2. All points above the curve represent states with $\Gamma_2 < 0$ (stripes) while all points below the curve represent

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