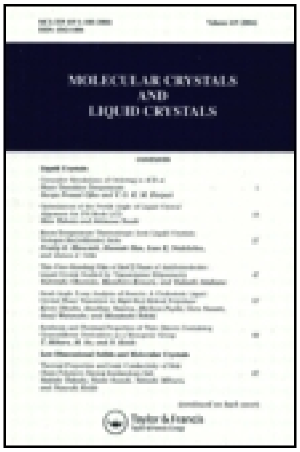

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Surface Transition in a Nematic Layer with Reverse Pretilt

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SURFACE TRANSITION IN A NEMATIC LAYER WITH REVERSE PRETILT

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Abstract Recently, a surface induced transition from homeotropic to planar alignment in a nematic layer and the corresponding optical response has been reported /K. Flatischler, L. Komitov, S. T. Lagerwall, B. Stebler, and A. Strigazzi, Mol. Cryst. Liq. Cryst., **198**, 119 (1991)/. In the case of surface conditions with reverse pretilt a similar transition from quasi-homeotropic bent alignment to quasi-planar splayed state can be obtained. As known, these two states are topologically incompatible, and therefore the transition is accompanied by nucleation of π-disclination lines, and even is reversible, it exhibits a big hysteresis. Moreover, it can be driven not only by temperature but also by an applied electric field, using a nematic with negative dielectric anisotropy. A simple theoretical description of such an alignment transition is presented.

INTRODUCTION

The interaction between a nematic liquid crystal and a solid substrate is a fascinating topic from fundamental and applied point of view, since the anchoring properties, yet far to be completely understood[1-6], can be a source of several electro-optical effects[7-10] studied in the last decade. Most of them can be directly utilized, for instance the so-called surface transition driven by temperature, which, starting from the pioneering work of Bouchiat et al.[11] ameliorated by Faetti et al.[12], was recently characterized by Köhler[13]. Last year we reported such a kind of surface transition occurring in a homeotropic nematic layer symmetrically anchored[14], giving also a phenomenological model, whereas the same phenomenon was later on being theoretically studied by Gabbasova et al.[15].

Moreover, Rosenblatt et al.[16] at the same time reported a transition from homeotropic to tilted alignment, giving also an overview on the state-of-the-art.

The aim of the present work is to describe the experimental evidence of the surface transition which takes place in a nematic layer with reverse pretilt, giving also a simple phenomenological model of the effect.

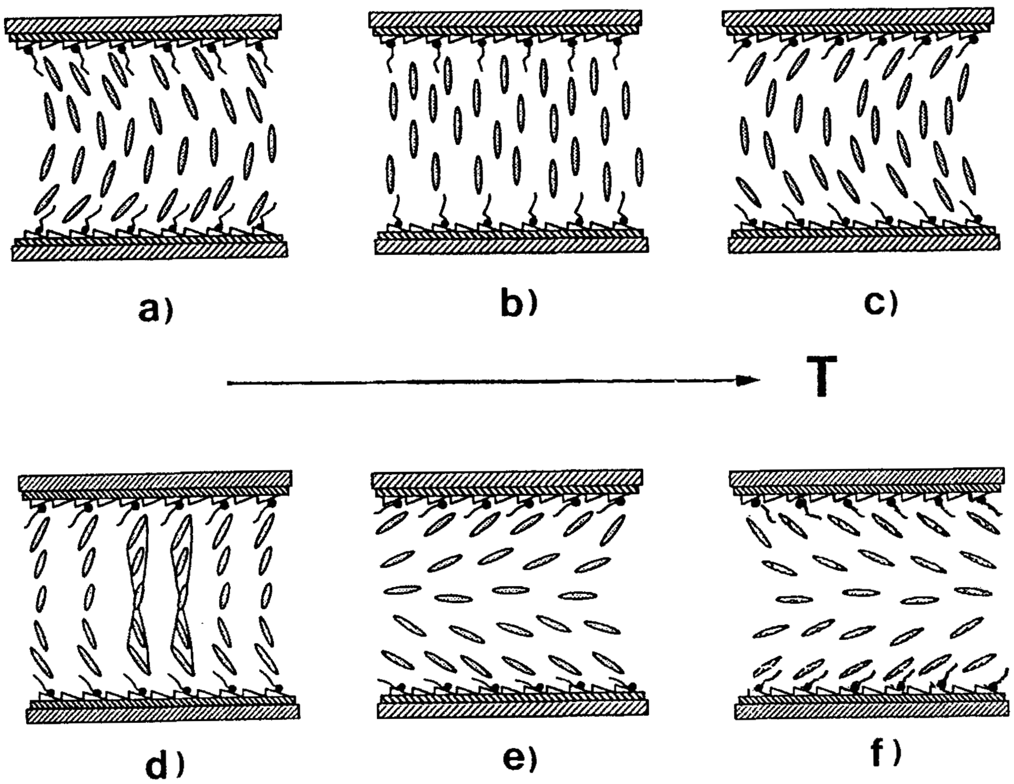


FIGURE 1 - Surface transition driven by temperature: **a)** initial bent state at room temperature T_a (bend I configuration); **b)** homeotropic alignment at $T_b > T_a$; **c)** bent state at $T_c > T_b$ (bend II configuration); **d)** π-twisted state at $T_d > T_c$; **e)** splayed state at T_e (splay I configuration) above the transition temperature (T_{cB} on heating and T_{cS} on cooling: the wide hysteresis provides $T_{cS} < T_{cB}$); **f)** splayed state (splay II configuration) which can be attained applying a destabilizing electric field to the bent state a).

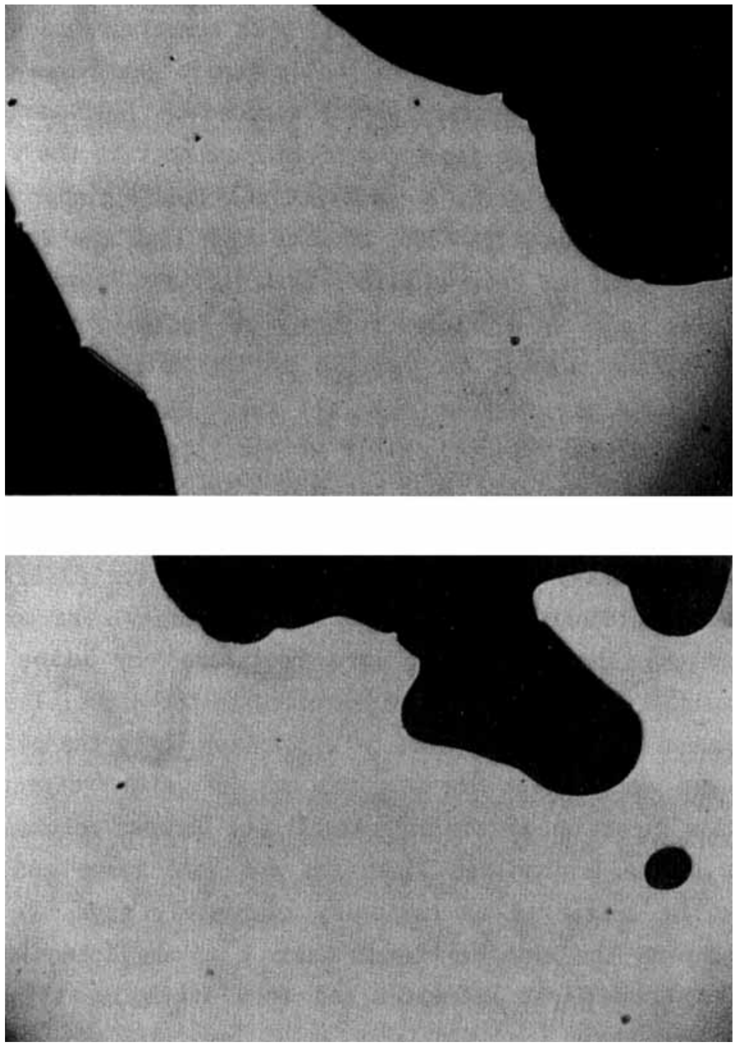
EXPERIMENTAL

A cell of conventional sandwich type with a gap of 18 m was used. The liquid crystal material ZLI 2806 (Merck) with nematic range -30.0°C to 99.5°C was filled into the cell in the isotropic phase. On the inner surfaces of the confining glass plates covered by ITO, an SiO_x film was obliquely deposited at angle ß = 5° with respect to the substrate. As known, such a layer gives a unidirectional quasi-planar alignment with molecular tilt about 25°-30°, on condition that the anchoring is weak. The SiO_x film was covered with a thin lecithin layer, by dipping the glass substrates in a diluted solution of lecithin in chloroform. The last treatment causes an increase of the molecular tilt (about 90°) at the substrate surface. The cell substrates were assembled in such a manner, that a reverse pretilt of the liquid crystal molecules at the confining surfaces was obtained. Thus, the nematic layer comprises an initial bend deformation as shown in figure 1-a.

The experimental cell is inserted in a Mettler FP52 hot stage with temperature controlled to within 0.1°C accuracy. The optical and electro-optical characteristics were registered by using a Zeiss Photomicroscope III Pol and the videorecording technique. The cell was placed between crossed polarizers in a position where the plane of the initial bend deformation is oriented at 45° with respect to the transmission direction of the polarizers. Due to bend deformation, the intensity of the transmitted light has non zero value and thus the cell does not appear to be optically completely dark. On heating, before reaching the critical temperature T_{cB}, the intensity of the transmitted light first decreases and then increases slightly. The critical temperature T_{cB}, similarly as T_c in /14/, was found to be about several degrees below the clearing point T_{NI}.

At T_{cB} a transition to another state, which is optically bright, begins at different points of the sample area and, following a slight rise in temperature, the new state fully occupates the whole sample (see figure 2). The transition was found to be reversible with a strong hysteresis: the critical temperature on cooling T_{cS} is lower than T_{cB}.

In order to define the changes of the alignment in the nematic



a)

b)

FIGURE 2 - Orthoscopic observations through crossed polarizers of the surface transition in a nematic layer with an initial bend deformation, upon heating. From a) to c) the temperature is increasing: a) T_1 = 93.2°C, b) T_2 = 93.6°C, and c) T_3 = 94.0°C. In the black areas the alignment is quasi-homeotropic (bent state), whereas in the bright yellow ones there is an induced splayed state. See Color Plate III.

c)

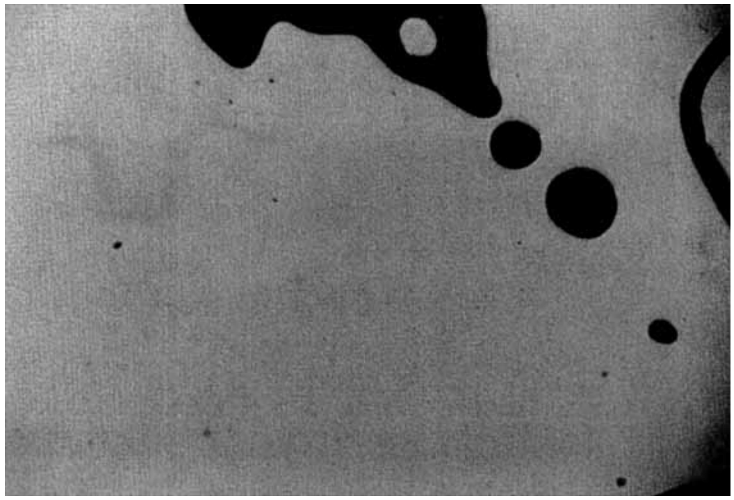


FIGURE 2 (continued) See Color Plate III.

layer when a surface transition is taking place, either at heating or at cooling, we performed conoscopic investigations. The results we obtained suggest the following picture of the transition schematically depicted in figure 1. On heating, before T_{cB} being reached, the initial bent state is transforming continuously with the temperature into an oppositely directed bent state via a homeotropic one. This transformation first causes decreasing and then increasing of the transmitted light intensity, the behavior we already mentioned above. On further heating, a transition to a splayed state via a π-twisted state takes place, as the temperature exceeds the critical one (see figure 1). Since there is a topological compatibility between homeotropic, bent and π-twist states, they can transform continuously from one to the other. However, these states are topologically incompatible with the splayed state, therefore the transition between them is discontinuous and is accompanied by nucleation of $|S| = 1/2$ disclination lines[17,18] (see figure 3). Due to that fact, the transition bend-splay exhibits a strong hysteresis, since an energy barrier has to be overcome.

As known, this transition can also be driven by an electric

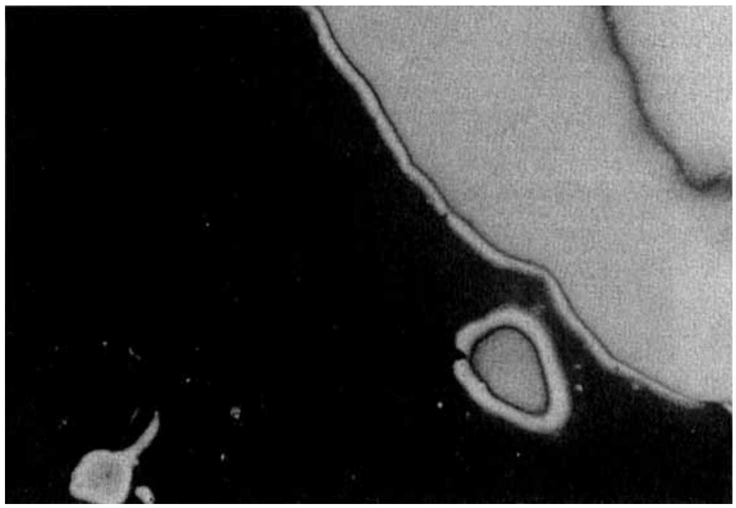


FIGURE 3 - Orthoscopic observation through crossed polarizers of the surface transition on cooling, at 91.8°C. The $|S|=\frac{1}{2}$ disclination lines separating the π-twist state from the splayed one are clearly visible. See Color Plate IV.

field[19]. In the presence of surface induced bend-splay transition the threshold voltage U_I for such a transition exhibits a strong temperature dependence (see figure 4). On heating, the threshold U_I has almost a constant value which rapidly decreases on approaching T_{cB} and becomes zero as T_{cB} is reached.

On cooling through a wide temperature range, the surface induced splayed state can be kept by an applied voltage U_K much lower than the one required for inducing the splayed state from the initial bend configuration (compare U_I and U_K in figure 4). The hysteresis character of the surface induced bend-splay transition could be useful for practical applications.

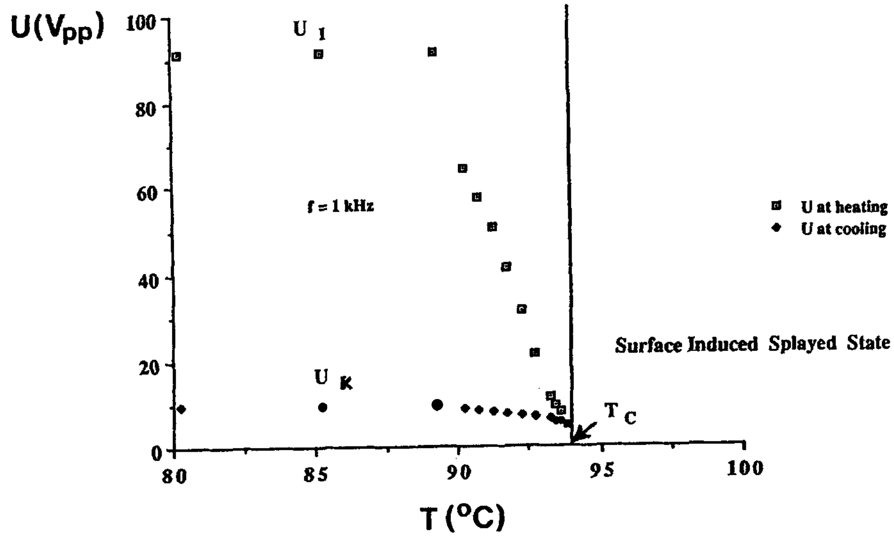
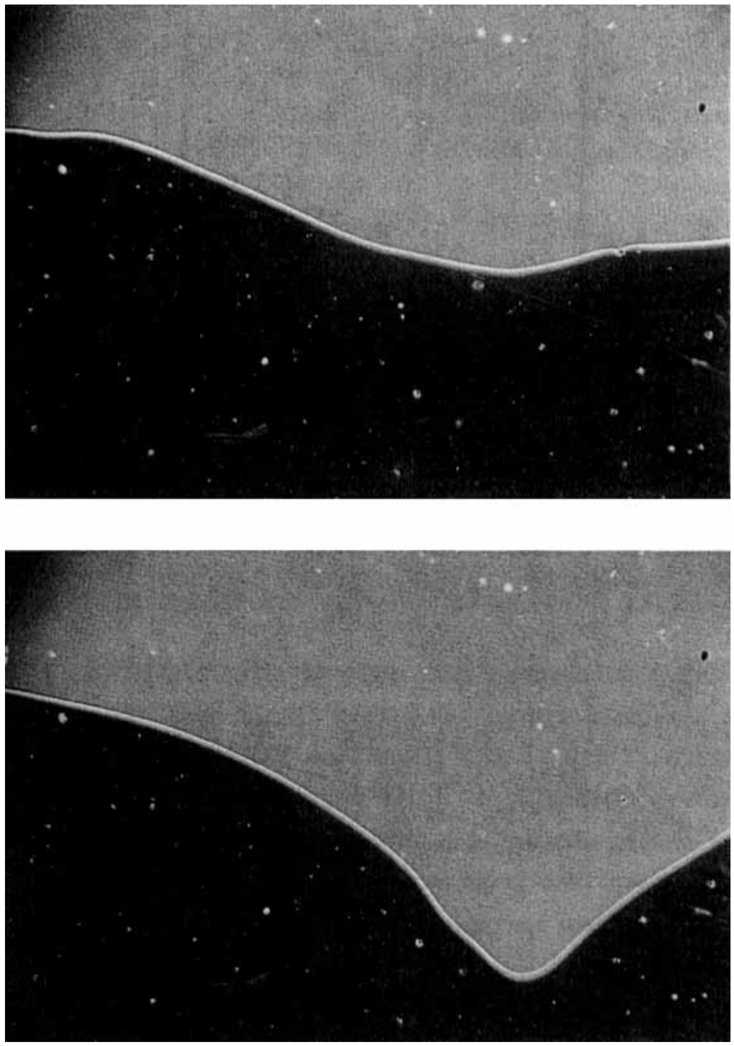


FIGURE 4 - Bend-splay transition: inducing voltage U_I and keeping voltage U_K as dependent on temperature T. Experimental data at low frequency (f = 1 kHz).

The microscope observations reported in figure 5 illustrate the surface alignment transition biased by the electric field at constant temperature (close to the transition point T_{cB}). In picture 5-a the effect of a voltage $U_a \ll U_I$ is shown: the black area corresponds to the bent state, whereas the pink area corresponds to the splayed state with tilt angle less pronounced than in the absence of the field. Note the bright stripes surrounding the black area, which is relevant to the π-twist state, separated through a long disclination line from the splayed state zone. Moreover, the bright spots, which are spread over the black area, are nucleation centers of splayed state zones. In picture 5-b ($U_a < U_b < U_I$) the area relevant to the splayed state is enlarged. But, on applying U_c of the order of U_I (see figure 5-c), the bent state comprises a very pronounced deformation, with high average tilt angle (bright green color area), whereas the splayed state area is further on enlarged and exhibits a smaller distortion.



a)

b)

FIGURE 5 - The effect of applied electric field on the surface transition (at increasing temperature) close to the critical temperature: in **a)**, **b)** the applied voltage is much lower than the inducing voltage U_I with $U_a < U_b$; whereas in **c)** U_c is almost equal to the inducing one. The pink (orange) colored areas in **a)**, **b)**, **c)** illustrate a splayed state more close to planar alignment, whereas the green colored area in **c)** is relevant to the strongly distorted bent state. See Color Plate V.

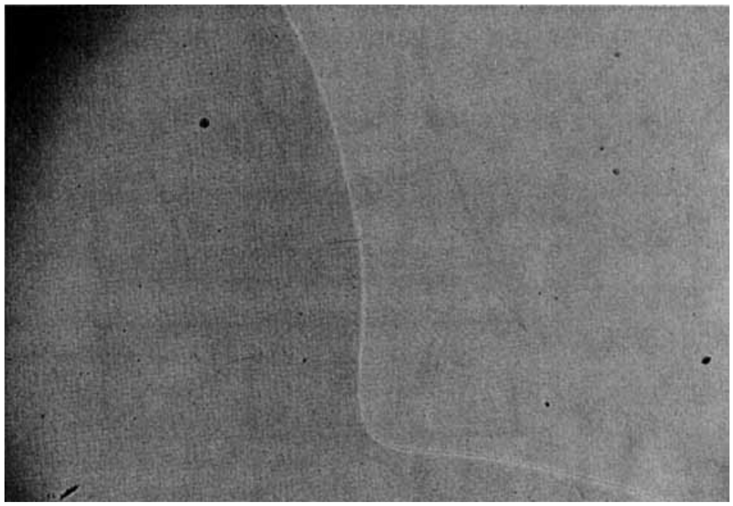
c)


FIGURE 5 (continued) See Color Plate V.

THEORETICAL

A nematic cell of thickness d is considered, with initial bent quasi-homeotropic alignment at room temperature (see figure 1-a). We assume anchoring conditions with reverse pretilt at the boundaries covered by SiO_x-aligning layers with sawtooth profile. Here the single SiO_x formation is oriented at an angle $\pm\alpha$ with respect to the substrates $z_0 = -d/2$ and $z_1 = d/2$, respectively, z being the co-ordinate normal to the substrates (see figure 6). Moreover, for the single SiO_x formation with sizes a and b, we also assume a ratio $a/b \ll 1$, i.e. the surface of size b is supposed to be mainly responsible for the alignment.

As in ref. /14/, two competitive easy directions, normal and parallel to the growing direction of the single SiO_x formation, are taken into account. Such easy directions are characterized by the anchoring strengths $W_H(T)$ and $W_P(T)$, respectively, which both depend on temperature[20,21]. For simplicity, we restrict ourselves to the hypotheses of one elastic constant K, and equal anchoring energies at both surfaces, and we assume $\alpha \cong \beta$.

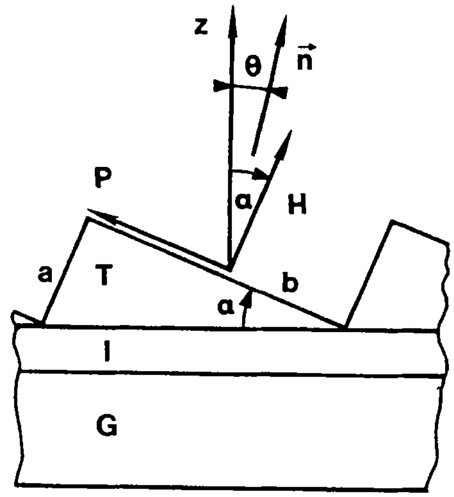


FIGURE 6 - Cross section of the cell substrate: G, glass; I, indium tin oxide; T, SiO_x teethlike formation. H and P are the two competing easy directions at the sawtooth surface relevant to the dimension b. The local nematic director **n** is defined by the tilt angle θ with respect to the substrate normal z.

Orientational Transition in the Absence of an Electric Field

In the low temperature range, the anchoring strength W_H is greater than W_P (see figure 7-a), thus the director profile **n**(z) presents a bend-splay deformation, being essentially bend.

In fact the reduced free energy of the layer is given by:

$$G = \int_{-d/2}^{d/2} \theta'^2 \, dz + (2/K) \left[W_H \sin^2 (\theta_1 - \alpha) + W_P \cos^2 (\theta_1 - \alpha)\right] \qquad (1)$$

where θ is the tilt angle with respect to the z-axis, the prime means the first derivative with respect to z, and θ_1 is the value assumed for θ at the upper surface (Note that $|\theta_1| \ll 1$ rad, since $\alpha \cong \beta \cong 5°$).

a)

b)

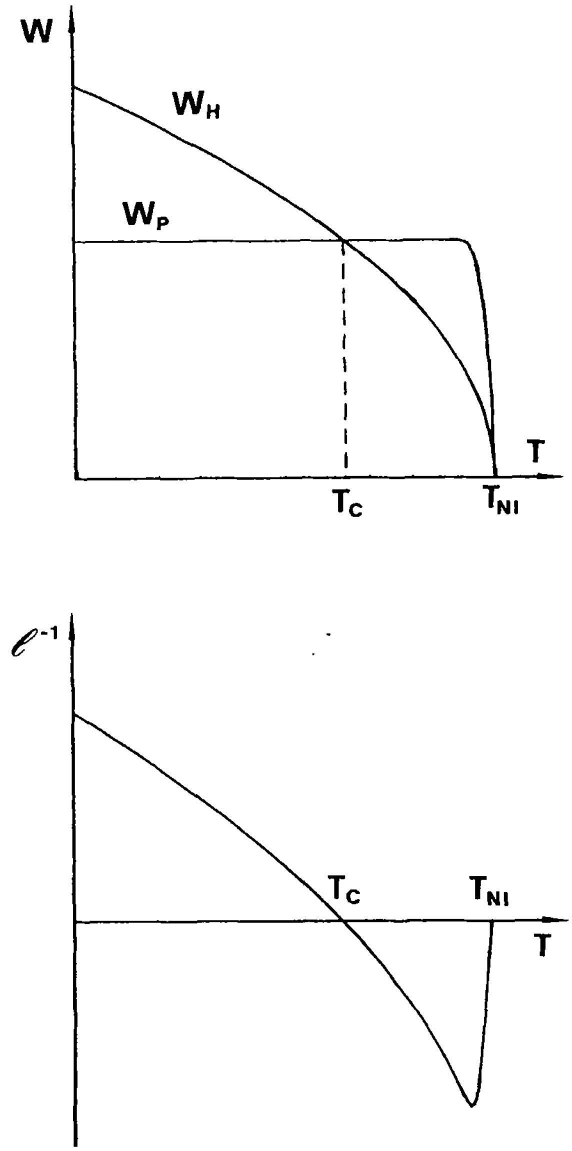


FIGURE 7 - Qualitative temperature T dependence a) of the anchoring strengths W_H, W_P for normal and parallel easy directions at the surfaces, respectively; b) of the reciprocal reduced extrapolation length ℓ. Note that $W_H \propto S^2$, W_P = const., and $K \propto S^2$, S being the scalar order parameter. T_{NI} is the clearing point (99.5 °C for ZLI 2806), T_c is the critical temperature for surface transition in the case of no pretilt (93.5°C).

Hence the Euler-Lagrange (EL) equation writes

$$\theta'' = 0 \tag{2}$$

with the boundary condition

$$2\,\theta'_1\,d + \ell^{-1}\sin 2\,(\theta_1 - \alpha) = 0 \tag{3}$$

$\ell \equiv K/[(W_H - W_P)d]$ being the reduced extrapolation length[22,23], which takes into account the balancing between the anchoring strengths connected with the two easy directions. Note that, according to the different behavior of W_H, W_P vs. temperature[24], ℓ can be also negative: in the case of symmetrical anchoring condition, the sign interchange does appear at the surface transition temperature T_c, several degrees below the clearing point[14].

Actually W_H is proportional to S^2 (S being the scalar order parameter), whereas $W_P \simeq$ const.: this means that, for a certain temperature T_c, $W_H = W_P$ is attained, giving zero value of the effective anchoring strength at the surfaces. In figure 7-b the temperature dependence of $\ell^{-1}(T)$ is depicted, taking into account the fact that K is also proportional to S^2.

The odd solution of eq. (2) reads

$$\theta = C_B\,z \tag{4}$$

where $C_B = 2\theta_1/d$. By inserting (4) into the boundary conditions (3) the transcendent equation

$$X = 4\,\ell\,\theta_1 = -\sin 2(\theta_1 - \alpha) = Y \tag{5}$$

is obtained. The actual value θ_1 is given by the proper intersection between the straight line $X(\theta_1)$ and the curve $Y(\theta_1)$. Such an intersection is shown in figure 8-a, as a function of the parameter ℓ, while in figure 8-b the function θ_1 vs. ℓ^{-1} is represented.

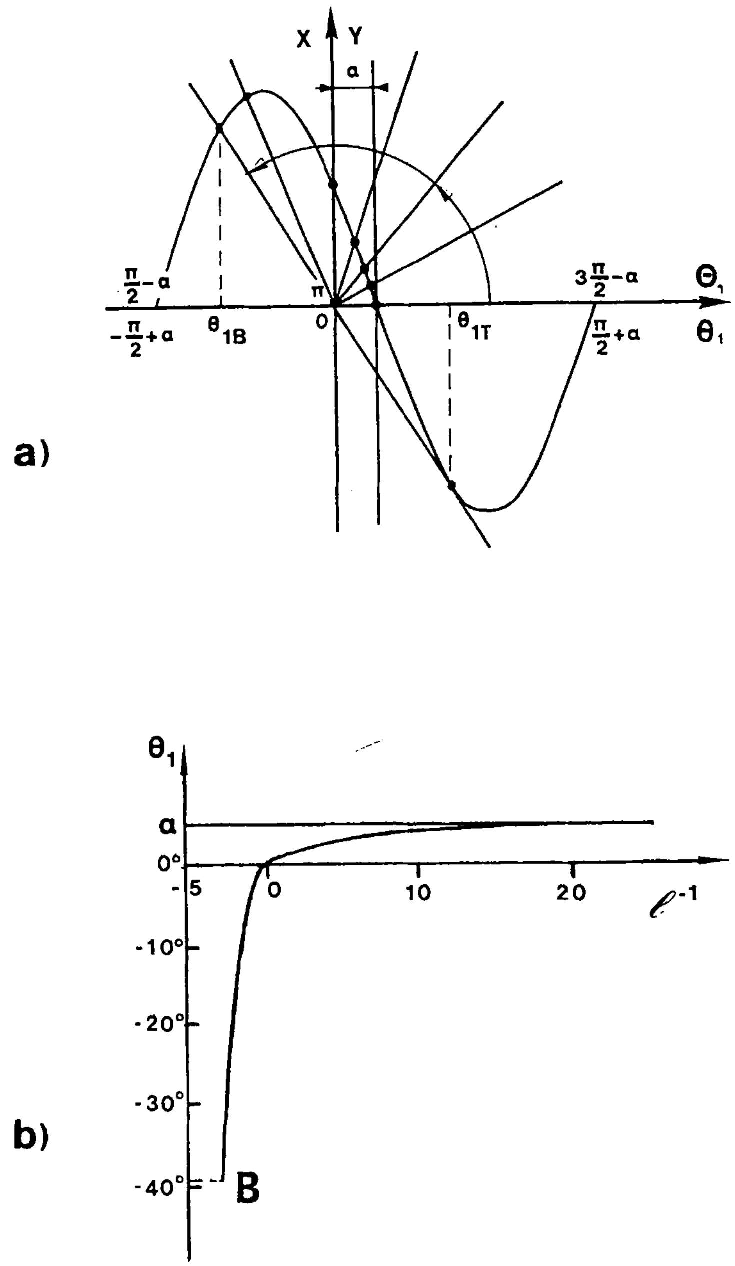


FIGURE 8 - a) Straight line $X(\theta_1;\ell)$ and curve $Y(\theta_1)$, giving b) the tilt angle at the upper substrate θ_1 vs. ℓ^{-1}. In the present case $\alpha = 5°$ is assumed. The point B gives the critical condition on heating.

In the present case the bend-splay structure becomes unstable when the straight line $X(\theta_1)$ is tangent to the curve $Y(\theta_1)$, this means when $\ell(T_{cB}) = -\cos 2(\theta_{1T} - \alpha)/2$, θ_{1T} characterizing the tangential point between the straight line and the curve. In our case $\theta_{1T} = 26.2°$, since $\theta_{1T} = \tan 2(\theta_{1T} - \alpha)/2$. Thus, the critical tilt angle $\theta_{1B} = -38.9°$ is reached when $\ell_B = -0.368$: i.e. at $T_{cB} > T_c$, upon heating.

The phenomenon of the surface transition occurring in nematic cells with reverse pretilt is characterized by a strong hysteresis. In fact, on approaching T_{cB} the distortion continuously transforms in a spatial one, since a mixed twist-splay-bend (π-twist) involves less energy than a planar deformation, allowing a diminishing of the bend-splay. On further heating, the mixed deformation collapses into a planar one, of mixed splay-bend, essentially characterized by splay. This implies the nucleation of π-disclination lines, since the bend-splay (bent state) and the splay-bend (splayed state) are topologically incompatible (see figure 9-a,b).

a)

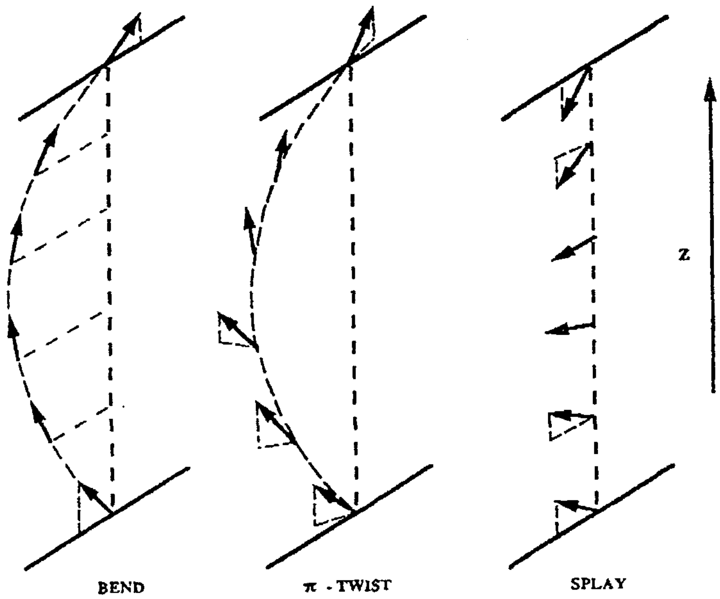
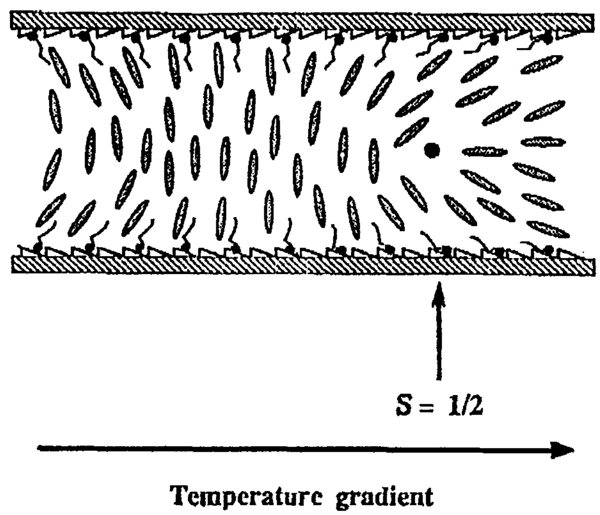


FIGURE 9 - a) Distortions involved in the surface transition in a nematic layer with reverse pretilt: bend, π-twist, and splay (from left to right). b) nucleation of π-disclination line in the surface transition driven by temperature.



b)

FIGURE 9 (continued)

By measuring the tilt angle $\Theta = \pi/2 - \theta$ from the x-axis parallel to the substrates, in the splayed state (configuration I, see figure 1-e) the Euler-Lagrange equation keeps the same form as in (2), with the solution

$$\Theta = C_S\, z + \pi \tag{6}$$

where $C_S = 2(\Theta_1 - \pi)/d$, which satisfies the boundary condition (3) with (ℓ^{-1}) replaced by $(-\ell^{-1})$. The same occurs in eq. (5).

Hence, on cooling the splayed structure becomes unstable just at the temperature T_{cS} when $\ell(T_{cS}) = -\ell(T_{cB})$, i.e. when $\Theta_{1S} = 141.1°$. As a consequence, $T_{cS} < T_c < T_{cB}$, i.e. the surface transition in the case of reverse pretilt will exhibit a large hysteresis.

Note that the predicted behavior of Θ_1 for the splayed state as a function of $-\ell^{-1}$ is the same as that depicted in figure 8-b, provided (ℓ^{-1}) is changed into $(-\ell^{-1})$, according to the previous description.

Orientational Transition in the Presence of an Electric Field

When an AC electric field perpendicular to the cell substrates is applied, either the homeotropic alignment is destabilized or the planar state is stabilized, owing to the negative dielectric anisotropy (ε_a = -4.0) of the nematic phase[22]. This means that, considering the bent state, the destabilizing term $g_E = (\xi d)^{-2} \sin^2\theta$ must be added in (1) to the bulk reduced free energy, where the reduced electric coherence length ξ is given by $\xi = (-4\pi K/\varepsilon_a)^{\frac{1}{2}}/(E\ d)$.

Hence in the absence of twist the EL equation reads:

$$2\ \theta'' d^2 + \xi^{-2} \sin 2\theta = 0 \tag{2'}$$

which is similar to the anharmonic pendulum equation.

Let us simplify the calculation, giving an approximate solution by linearizing eq. (2'). This convenience is acceptable until $|2\theta| < 45°$ everywhere in the cell, if a precision less than 11% on the local tilt angle is considered as sufficient.

The linearized EL equation reads

$$\theta'' d^2 + \xi^{-2}\ \theta = 0 \tag{7}$$

with the linearized boundary condition:

$$\theta'_1 d + \ell^{-1}(\theta_1 - \alpha) = 0 \tag{8}$$

Eq. (7) is satisfied by the odd solution

$$\theta = c_B \sin(z/\ \xi\ d) \tag{9}$$

where $c_B = \theta_1/\sin(1/2\ \xi)$. By inserting (9) into (8) we obtain

$$\theta_1 = \alpha/\left[1 + \ell(1/\ \xi)\ \text{cotan}\ (1/2\xi)\right] \tag{10}$$

which gives for the destabilization of the bent state the approximate threshold

$$\ell_B = -\ \xi \tan(1/2\xi) \tag{11}$$

On the other hand, considering the splayed state we have

$$\psi'' d^2 - \xi^{-2}\psi = 0 \tag{7'}$$

and

$$\psi'_1 d - \ell^{-1}(\psi_1 - \alpha) = 0 \tag{8'}$$

instead of the EL equation (7) and of the boundary condition (8), in the same hypothesis, with $\psi = \Theta - \pi$ and $\psi = \Theta$ in the cases of I and II configuration, respectively (see figure 1-e,f).

This means that,

$$\psi = c_S \, \mathrm{sh}(z/\xi d) \tag{12}$$

being the solution of (7'), the approximate threshold writes

$$\ell_S = \xi \, \mathrm{th}(1/2\xi) \tag{13}$$

In figure 10 the approximate critical values of the reciprocal reduced extrapolation lengths ℓ_B^{-1}, ℓ_S^{-1} are shown, being dependent on the effective anchoring strength $(W_H - W_P)$, as functions of the reciprocal reduced coherence length ξ^{-1}, which is proportional to the applied field E.

DISCUSSION

The surface transition, described in the previous section, behaves in an essentially different way compared to the one recently reported in reference /14/. This is mainly due to the fact that a symmetrical pretilt $\theta_0 = \theta_1$ gives uniform alignment in the nematic layer, whereas a reverse pretilt $\theta_0 = -\theta_1$ always causes a distortion. In the later case, in the low temperature range (i.e. at temperature much below the clearing point) this implies an initial quasi-homeotropic state revealing essentially a bent deformation.

The surface transition discussed in reference /14/, where a symmetrical pretilt was considered, is smooth, practically without

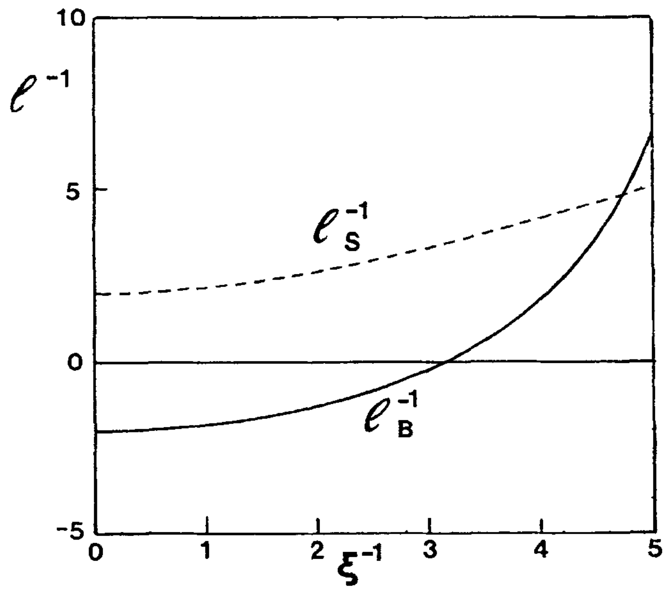


FIGURE 10 - Approximate critical values of the reciprocal reduced extrapolation length ℓ^{-1} in the presence of an electric field E vs. the reciprocal electric coherence length ξ^{-1}, which is proportional to E. Note that ℓ_B and ℓ_S are relevant to the bent and splayed state, respectively. The curves show that when the field is applied in the bent state, the surface transition is favored, whereas the splayed state is stabilized.

hysteresis, and does not require a nucleation of disclination lines, since the two equilibrium states below and above the critical temperature T_c are topologically compatible; the transition is expected to be of second order. Instead, in the present paper we are dealing with a surface transition which exhibits a strong hysteresis, and formation of π-disclination lines, owing to the topological incompatibility of the initial bent state with the final splayed state: thus the transition temperature on heating (T_{cB}) is greater than the transition temperature on cooling (T_{cS}). The transition is expected to be of first order.

Such a behavior was found experimentally (see figures 2, 3) and described by means of a simple phenomenological model, taking into account the competition between two easy directions[25,26], the first normal and the second parallel to the surfaces of the SiO_x teeth (see figures 6, 7). The behavior predicted by the model (see figure 8) is in good agreement with the experimental observations.

The presence of an electric field E normal to the cell substrates deeply affects the surface transition, since the bent state is destabilized, whereas the splayed state is found to be favored, when the dielectric anisotropy of the nematic is negative: compare figures 4 and 5, illustrating the experiment, with figure 10, showing the model predictions. In particular, it is predicted that on applying a low electric field (for instance giving $\xi^{-1} = 2$) when the equilibrium state of the nematic layer is the bent one, the surface transition temperature $T_{cB}(\xi^{-1})$ turns out to be shifted towards lower temperatures. In fact, $T_{cB}(\xi^{-1}) < T_{cB}(0)$ is obtained, since $\ell_B^{-1}(0) < \ell_B^{-1}(\xi^{-1}) < 0$. By increasing the field (for instance $\xi^{-1} = 4$), a greater shift of temperature transition is achieved, since $\ell_B^{-1}(\xi^{-1}) > 0$; hence $T_{cB}(\xi^{-1}) < T_c$. Moreover, applying the field when the splayed state is already attained, the field itself prevents the reverse surface transition from splayed to bent state, which may arise at temperatures well below T_c, always resulting in $\ell_S^{-1} > 0$.

CONCLUSION

A new kind of surface transition driven by temperature and biased by an electric field is reported. Such a transition takes place in nematic layers with pretilt reversal several degrees below the clearing point, and exhibits strong hysteresis. Due to its features, the alignment transition leads to certain changes in the optical properties of the liquid crystal layers. These are attractive from a fundamental point of view, and may be useful for different applications.

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