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Polar anchoring energy and order parameter at a nematic liquid crystal-wall interface

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By measuring the integrated birefringence versus thickness of homogeneous and distorted nematic liquid crystal configurations, we have obtained the anchoring energy and the change of the order parameter at the orientating layer. The measurements were made for planar and tilted orientating SiO layers and for rubbed polyimide. The tilted orientating SiO layer shows the most significant decrease of the order parameter near the interface, which can be explained by its microscopically inhomogeneous alignment.

1. Introduction

The anchoring strength of a liquid crystal at an orientating layer is characterized by an anchoring energy or equivalently by an extrapolation length L , where the ratio of L to the cell thickness determines the influence of the anchoring properties on the director configuration in the cell. Different methods have been developed to measure the extrapolation length. They use either a field induced director distortion (for example [1]) or a distortion caused by different tilt angles at the two boundaries (for example [2]) to induce a torque field at the interface. For a pure elastic deformation the torque at the boundary is increased by a decreasing cell thickness and has only a significant influence on the boundary tilt angle if the cell thickness is of the order of magnitude of the extrapolation length. For relatively low values of the extrapolation length the methods without an external field therefore require measurements for very thin cells, where additional complications may result from a disordered surface layer with a thickness of the same or even larger order of magnitude than the extrapolation length. This point will be explained later. On the other hand, the torque at the boundary can be increased by applying an external field which enables measurement of low values of the extrapolation length for larger cell thicknesses.

Measured values for L are, for example, $0.065 \mu\text{m}$ for planar orientating SiO [1], but also $1 \mu\text{m}$ [2] for a rubbed silane surface. It is evident therefore that the influence of the anchoring properties may become more important for thin liquid crystal cells.

Recently, Yokoyama *et al.* [3] found from measurements of the temperature dependence of the anchoring strength that a reduced surface order parameter should contribute to the extrapolation length. Experimental evidence for a reduced surface order was obtained from birefringence measurements on planar orientating SiO [4].

2. Theory

The polar anchoring of the liquid crystal molecules at an orientating layer can be described in the framework of continuum theory [5] in terms of an interfacial free energy function, $\gamma(\Theta_0 - \Phi_0)$, where Θ_0 is the tilt angle at the interface and Φ_0 is its value for a homogeneous director orientation (the preferred orientation). The

interfacial energy gives the amount of work necessary to turn the director orientation at the interface away from its preferred direction. For small deviations ($\Theta_0 - \Phi_0$) we can expand the interfacial energy to give

$$\gamma(\Theta_0 - \Phi_0) \approx \text{constant} + \frac{1}{2}E_a(\Theta_0 - \Phi_0)^2, \quad (1)$$

where the linear term is assumed to disappear for symmetry reasons. The material parameter E_a is called the anchoring energy.

We consider an untwisted director configuration depending only on the coordinate z normal to the orientating layers. A dimensionless coordinate is defined as $\xi = z/d$, where d is the thickness of the cell. By minimizing the total free energy we obtain the well-known Euler–Lagrange differential equation [6] for the tilt angle through the cell $\Theta(\xi)$ and an additional equation, describing the torque balance at the boundary

$$(1 + K \sin^2 \Theta) \left. \frac{d\Theta}{d\xi} \right|_{\xi=0,1} = \frac{d}{K_1} \frac{\partial \gamma}{\partial \Theta_0} \approx \frac{d}{L} (\Theta_0 - \Phi_0), \quad (2)$$

$K = K_3/K_1 - 1$, where K_1 and K_3 are the splay and bend elastic constants. Here we have defined an extrapolation length L as $L = K_1/E_a$. The ratio of the cell thickness to the extrapolation length determines the tilt angle at the boundary.

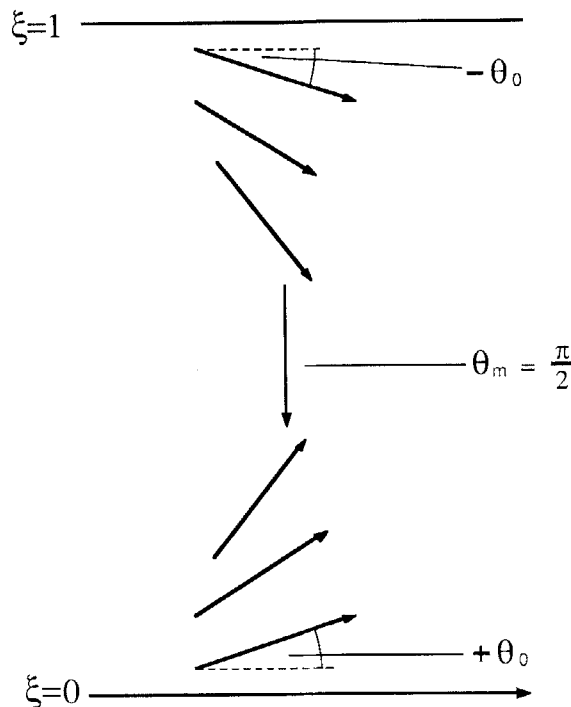


Figure 1. Director configuration produced by opposite tilt angles at the two boundaries.

For our method we have used the director configuration shown in figure 1. An antisymmetric director orientation relative to the centre of the cell is caused by opposite tilt angles at the two boundaries. With a sufficiently high external field the configuration is stable for every non-zero value of the tilt angle. Independently of the cell thickness, the orientation is exactly homeotropic in the centre of the cell and for

voltages above three times the threshold voltage (for ZLI1132, from Merck) the derivative $d\Theta/d\xi$ in the centre of the cell is very close to zero; both simplify the calculation of the director orientation. In the region $\xi = 0$ to $\xi = \frac{1}{2}$ and $\xi = 1$ to $\xi = \frac{1}{2}$ the director configuration is equivalent to a hybrid aligned nematic (HAN) [2] cell with thickness $d/2$ and a fixed homeotropic boundary orientation.

Our method for determining the anchoring energy is based on the fact that for constant (or zero) voltage across the cell and for a fixed boundary tilt angle Θ_0 the function $\Theta(\xi)$ is independent of the cell thickness. Measuring the integrated birefringence, defined as

$$\Delta l = \frac{2\pi d}{\lambda} \left(\int_0^1 \frac{n_e d\xi}{[1 + v \sin^2 \Theta(\xi)]^{1/2}} - n_o \right), \tag{3}$$

where $v = (n_e/n_o)^2 - 1$, the curve Δl versus d should be a straight line intersecting the d axis at the origin, provided that the tilt angle at the boundary is independent of the cell thickness. As can be seen from equation (2), the deviation ($\Theta_0 - \Phi_0$) increases with decreasing cell thickness. In our director configuration the tilt angle tends to become homeotropic for low cell thicknesses, so that the slope $\partial\Delta l/\partial d$ decreases. When $d/L \gg 1$, Θ_0 tends to its preferred direction and $\Delta l(d)$ becomes a straight line of the form

$$\lim_{d/L \rightarrow \infty} \Delta l(d) = a(d - L^*). \tag{4}$$

For zero voltage the function $\Delta l(d)$ is the same as for the HAN cell if L is replaced by $2L$. The parameters a and L^* for this case are given explicitly in [2]. With a voltage applied we found an explicit formula for a and L^* with the assumption that $|d\theta(\frac{1}{2})/d\xi| \ll 1$ which is indeed satisfied as noted previously. The parameters are of the form

$$a = f_1(\Phi_0, K, \gamma, v, K_1, \varepsilon_{\perp}, n_e, U), \tag{5}$$

$$L^* = Lf_2(\Phi_0, K, \gamma, v, K_1, \varepsilon_{\perp}, n_e, U), \tag{6}$$

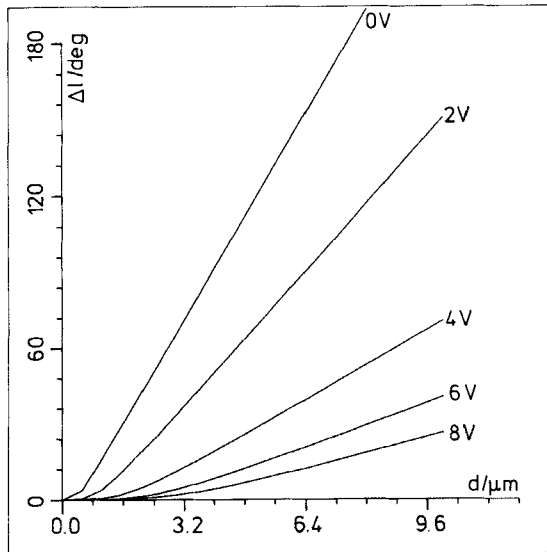


Figure 2. Integrated birefringence, Δl , versus cell thickness d calculated for voltages of 0, 2, 4, 6 and 8 V, with $L = 0.1 \mu\text{m}$.

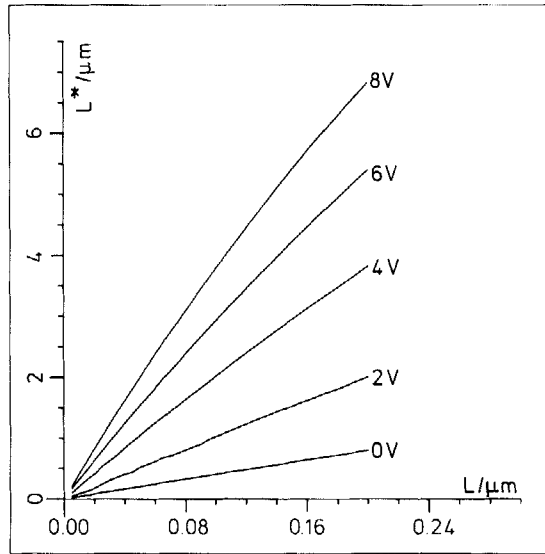


Figure 3. Dependence of the intersection point of the $\Delta l(d)$ asymptote L^* , on the extrapolation length L calculated for voltages of 0, 2, 4, 6 and 8 V.

where γ and ν are the dielectric and optic anisotropy, respectively, and U is the voltage across the cell. The point of intersection L^* is proportional to the extrapolation length L whereas the slope a does not depend on it. The functions f_1 and f_2 are rather complicated and involve integrals that can be evaluated only numerically. We have therefore performed all calculations with the programs described in [7].

Figure 2 shows the calculated curves for ZLI 1132 ($K_3/K_1 = 2.42$, $\Delta\epsilon = 10.3$, $\Delta n = 0.137$) and a preferred director orientation of 19° . In figure 3 the dependence of L^* on the extrapolation length for several voltages is shown. The preferred director orientation at the boundary, Φ_0 , can be determined from the measured slope a . Moreover, the functional dependence of $\gamma(\Theta_0)$ can be calculated from the measured integrated birefringence with the help of equations (2) and (3) and the Euler-Lagrange equation for $\theta(\xi)$, but this is not our aim here.

Measuring Δl versus d for a homogeneous director configuration $\Theta(\xi) = \Theta_0$ (i.e. the same tilt angle at both boundaries), the result should be a straight line intersecting the origin. We have observed, experimentally, for some orientating layers the extrapolation of the straight line $\Delta l(d)$ intersecting the origin at a finite thickness d^* in the region of $0.3 \mu\text{m}$. This indicates a reduction of birefringence and, therefore, of the order parameter near the interface [4]. To take this effect into account, we assume the simple model of a totally disordered layer with thickness $d^*/2$ at the interface. In this model the effect of the reduced surface order is simply a reduction of the cell thickness by an amount d^* . This holds even for a distorted director configuration, as can be seen from equation (3), if we assume that the director profile $\theta(\xi)$ is not influenced by the disordered layer. The integrated birefringence Δl is therefore reduced by a factor $(d - d^*)/d$.

3. Experiment

We have used wedge shaped cells to vary the cell thickness. Balzers glass substrates of thickness 3 mm coated with conductive transparent electrodes (indium-tin oxide) where cleaned by the usual methods. Three kinds of orientating layers were used.

(a) An approximately 150 Å thick SiO layer was vacuum evaporated at an angle of incidence of 60° from the normal, which is known to give a planar orientation with the director oriented perpendicular to the plane of incidence.

(b) An SiO layer of the same thickness was vacuum evaporated at an angle of 85° from the normal, resulting in a tilt angle of between 15° and 30°, with the director being in the plane of incidence.

(c) An approximately 500 Å thick polyimide film was spin coated on the substrates. Homogeneous alignment was achieved by unidirectional rubbing, resulting in a tilt angle of between 0 and 5°, dependent on the rubbing procedure, with a director orientation parallel to the rubbing direction.

The two glass substrates were separated at one end by a 10 μm Mylar spacer and they were in direct contact at the other end to form a wedge. For the cells with a distorted director orientation the electrodes were etched to prevent electrical contact at the thin end of the cell. To ensure the condition of constant voltage across the cell, the overlapping area of the two electrodes was relatively narrow and also the two substrates overlapped only in a small area, so that the electrodes outside could be coated with electrically conductive adhesive and the electrical resistance was minimized.

The thickness of the wedge shaped cells was measured at about 30 equidistant points before filling, using an optical interference method. The cell was mounted on a z - ϕ stage and the transmittance was measured for a variable angle of incidence using monochromatic light from a He-Ne laser. From the extrema of the transmittance versus the angle of incidence curve the cell thickness can be determined. By exact measurement of the transmittance through the cell and careful (automatic) evaluation, the error could be kept below 1 per cent. With the same apparatus the integrated birefringence, that is the phase difference between the extraordinary and the ordinary ray through the cell, was measured after filling. We have used the rotating analyser method as described in [9], where the phase angle was determined by a lock-in amplifier.

Filling was performed at room temperature with the nematic phase ZLI 1132 from Merck, and good alignment was verified using a polarizing microscope. The tilt angle of the cells with homogeneous director orientation was measured by the improved magneto-optic null method [8]. For the orientating layer (a) the tilt angle at the boundary was 0.36°, for (b) 19° and 23.2°, and for (c) 1.8°.

We determined the birefringence Δn of ZLI 1132 at 20°C and a wavelength of 632.8 nm to be 0.137 from the slope of the $\Delta l(d)$ curve for the cell with a homogeneous orientation and the values of the tilt angle measurement. For distorted director configurations we calculated the preferred director orientation Φ_0 with this result and the slope of the $\Delta l(d)$ asymptote without the applied field. Good agreement with the results from the magneto-optic measurements was found.

4. Results

We consider first the measurements for the cells with homogeneous orientation. The results of the measurements for the planar orientating SiO, tilted orientating SiO and the rubbed polyimide surface are shown in figures 4, 5 and 6, respectively. The tilted orientating SiO layer shows the most significant decrease of the order parameter. The intersection point of the $\Delta l(d)$ asymptote is at a d^* of 0.3 μm corresponding to an inactive surface layer thickness of 0.15 μm. For the planar

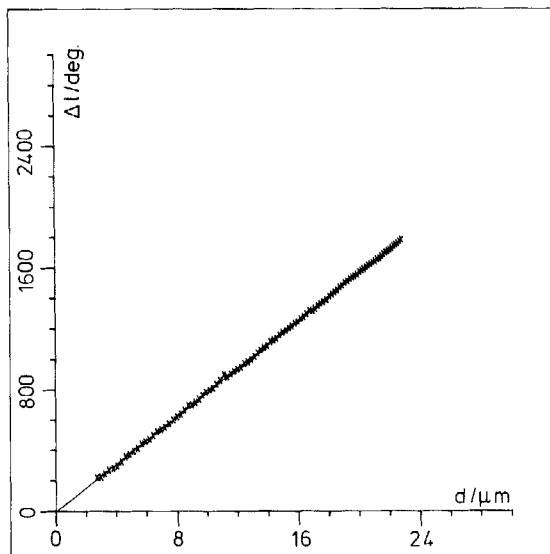


Figure 4. Integrated birefringence versus cell thickness measured for ZLI 1132 on planar orientating SiO, tilt angle 0.37° and a homogeneous director configuration.

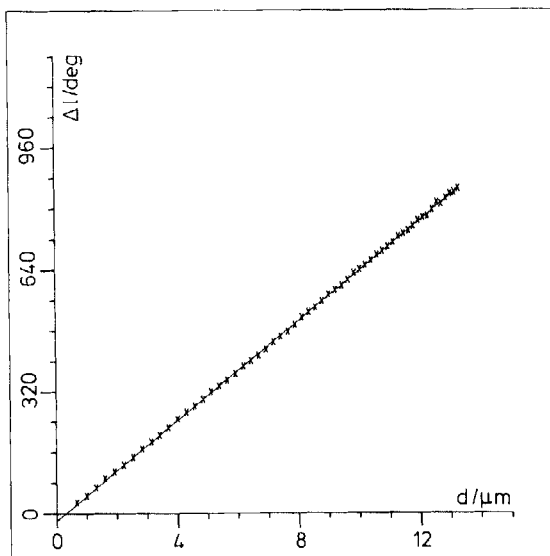


Figure 5. Integrated birefringence versus cell thickness measured for ZLI 1132 on tilted orientating SiO, tilt angle 23.5° .

orientating SiO, d^* is nearly one order of magnitude lower. An approximate value for a d^* of $0.03 \mu\text{m}$ is given by our measurements. This is contrary to the results of Wu and Efron [4] for the nematic phase E7 on planar orientating SiO. By measuring the integrated birefringence of several cells of different thickness they found a value of $0.38 \mu\text{m}$ for the intersection point d^* .

The difference in the thickness of the disordered surface layer between planar and tilted orientating SiO can be understood qualitatively by considering the two different

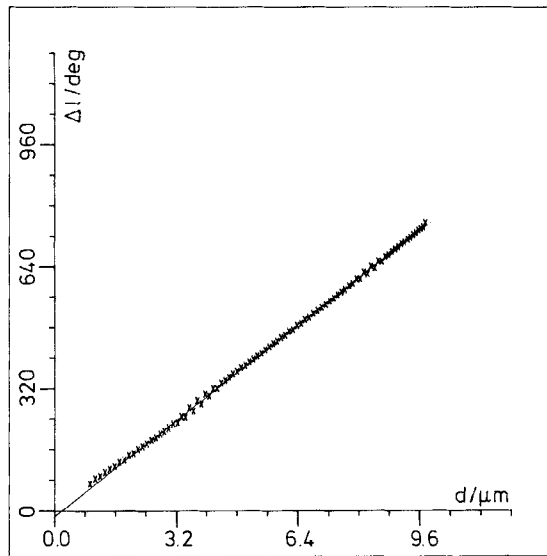


Figure 6. Integrated birefringence versus cell thickness measured for ZLI 1132 on rubbed polyimide layer, tilt angle 1.8° .

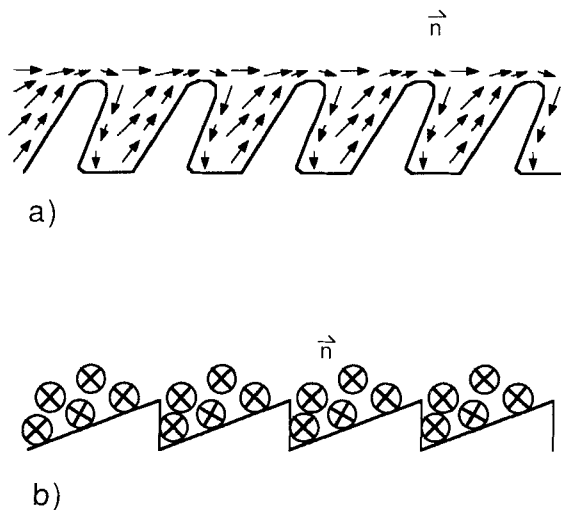


Figure 7. Assumed director orientation at (a) the tilted orientating SiO layer and (b) the planar orientating SiO layer.

aligning mechanisms. Both kinds of vacuum evaporation are known to produce some kind of grooved surface. For tilted orientating SiO produced by evaporation with an angle of 85° from the normal the director is orientated perpendicular to the grooves (see figure 7(a)). This causes a large director distortion at the interface because of the irregular shape of the surface. The orientation will be homogeneous at some distance from the surface. For planar orientating SiO produced by evaporation with an angle of 60° from the normal, the director is orientated parallel to the grooves (see figure 7(b)) and no director distortion is induced by the grooved surface. However, the amplitude of the grooves is about 100 \AA and therefore more than one order of

magnitude lower than the thickness of the disordered layer, so this model can only give a qualitative explanation.

The intersection point, d^* , for the rubbed polyimide layer was determined to be $0.12 \mu\text{m}$, which lies between the values for the two different SiO layers.

As a consequence of this result the decrease of the surface order parameter has to be considered whenever dealing with ultrathin cells, e.g. in all experiments for the determination of the anchoring energy where a wedge geometry is used. However, it is not quite clear how this can be done, because the functional dependence of the order parameter and also the influence of an external field on the order parameter is unknown.

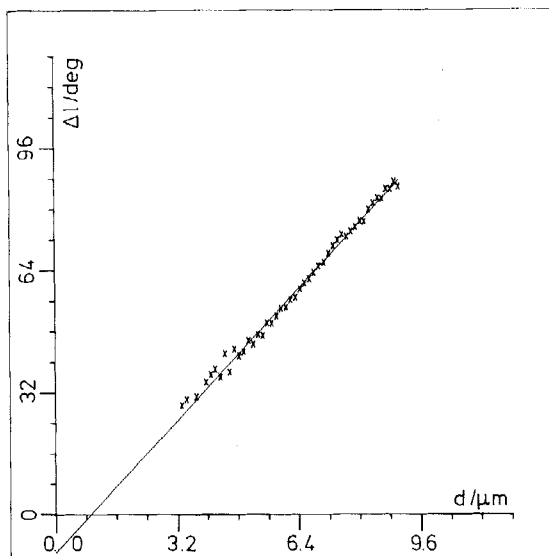


Figure 8. Integrated birefringence versus cell thickness measured for the distorted director configuration; applied voltage 3.42 V.

For the calculation of the anchoring energy from the intersection point L^* we assume the simple disordered layer model to a first approximation. We therefore have to correct L^* by

$$L_{\text{corr}}^* = L^* - d^*. \quad (7)$$

Figure 8 shows the measured $\Delta l(d)$ curve for a cell with a distorted director configuration. With the corrected point of intersection, we calculate an extrapolation length, L , for ZLI 1132 on tilted orientating SiO to be 350 \AA , which is approximately half of the value measured by Yokoyama and van Sprang [1] for 5 CB on planar orientating SiO; for tilted SiO no relative values were available. With $K_1 = 8.67 \times 10^{-12} \text{ N}$ we obtain for the anchoring energy, E_a , a value of $2.5 \times 10^{-4} \text{ J m}^{-2}$. To clarify the influence of the reduced surface order on the extrapolation length further, more measurements have to be made, especially on parallel orientating SiO, to compare it with these results. Moreover additional measurements at high applied fields should be carried out, where the intersection point L^* is much larger than the thickness of the disordered layer.

In order to see the influence of such relatively strong anchoring for practical applications we have calculated the mid-plane tilt angle versus voltage characteristic

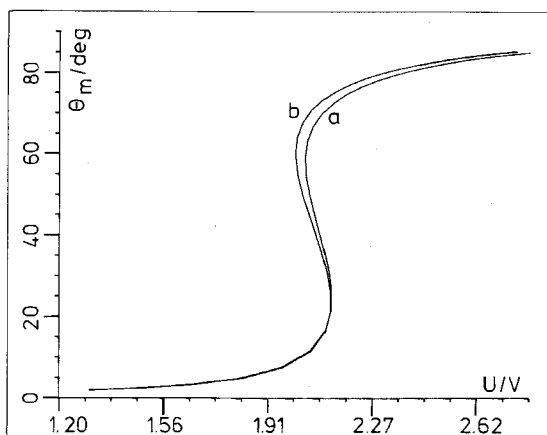


Figure 9. Mid-plane tilt angle versus applied voltage for an SBE cell calculated for with an extrapolation length (a) $L = 0$ and (b) $L = 350 \text{ \AA}$. Material parameters for ZLI 1132, total twist 270° , cell thickness $6.5 \mu\text{m}$.

for a 270° twisted (SBE) [10] cell using the material parameters of ZLI 1132. For the functional dependence of the interfacial energy we have assumed

$$\gamma(\Theta_0) = \frac{1}{2} E_a \sin^2(\Theta_0 - \Phi_0).$$

The results for $E_a = \infty$ and $E_a = 2.5 \times 10^{-4} \text{ J m}^{-2}$ are shown in figure 9. The hysteresis width [11] is increased from 0.092 to 0.124 V by this value of the anchoring energy.

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