Determination of liquid crystal director pretilt angle with respect to the orienting surface using optical phase shift

N. M. Shtykov, V. V. Lazarev, and M. I. Barnik

Research Institute of Organic Intermediate Products and Dyes, 103787 Moscow, Russia

A technique using liquid crystals whose dielectric anisotropy is an alternating function of frequency has been proposed for determination of liquid-crystal director pretilt angles with respect to orienting surfaces. The pretilt angle is derived from measurements of the optical phase difference in electric fields with frequencies above and below the anisotropy inversion frequency. The measured pretilt angles range between 0 and 90°, and the measurement uncertainty is the lowest near the middle of this interval. © *1996 American Institute of Physics.*
[S1063-7761(96)02411-0]

1. INTRODUCTION

The angle between the director of a nematic liquid crystal and the orienting surface, which is commonly called the pretilt angle, has a considerable effect on electro-optical characteristics (voltage–contrast, temporal, viewing angle, etc.) of liquid-crystal display devices. Therefore an accurate and reliable technique for measuring the pretilt angle is needed to optimize the parameters of such displays, for research and development of new orienting plates, and methods for preparing orienting surfaces.

There are several techniques for measuring the pretilt angle, the most popular of which are crystal rotation[1,2] and various versions of the magnetic technique.[3-5] All of them, however, have their flaws. In the crystal-rotation technique, the tolerance on the liquid-crystal thickness is very fine, the range of measured angles is quite narrow (0–13° and 77–90°), and the procedure of computer-aided fitting of the phase delay versus turning angle of the liquid-crystal cell is complicated. In the magnetic technique, the measuring facility is rather sophisticated and the capacitance of the liquid-crystal cell is an unstable parameter when the magnetic field is aligned with the director at the orienting surface.

The aim of this work was to develop a new technique for measuring the pretilt angle of a liquid crystal at an orienting surface. The measured parameter is the optical phase difference as a function of voltage at two frequencies, and the method exploits the property that in some liquid crystals, the dielectric anisotropy is an alternating function of frequency. In such liquid crystals,[6,7] the dielectric constant ε_p measured along the director at low frequency is larger than the dielectric constant ε_n with respect to the electric field perpendicular to the director, and the dielectric anisotropy $\Delta\varepsilon = \varepsilon_p - \varepsilon_n$ is positive. At a certain frequency f_0, the dielectric constants are equal and $\Delta\varepsilon = 0$; at frequencies higher than f_0, the dielectric constant along the director, ε_p, is lower than ε_n, and the dielectric anisotropy $\Delta\varepsilon$ is negative. In order to calculate the pretilt angle, one must determine the phase difference in the initial state of the liquid crystal with its pretilt angle, and the total phase delay in the layer with the planar alignment at zero tilt angle. Additional parameters needed for the calculation are the refractive indices of the

liquid crystal, which are determined independently.

2. THEORETICAL BACKGROUND OF THE TECHNIQUE

An oriented layer of a liquid crystal between two plates, on which the directors are parallel, is a uniaxial crystal with the optical axis directed with respect to the plates at the same angle as the liquid-crystal director (Fig. 1a). In a low-frequency electric field with $f_{LF} < f_0$, where f_0 is the frequency at which the sign of $\Delta\varepsilon$ is inverted, the Frederics effect takes place if $0 \leqslant \theta_t < 90°$. As a result, the liquid-crystal director and, consequently, its optical axis are aligned with the electric field, i.e. perpendicular to the orienting surfaces, except in thin layers near the surfaces (Fig. 1b), where the director has its initial alignment. When a high-frequency voltage ($f_{HF} > f_0$ and $\Delta\varepsilon < 0$) is fed to the cell, and $0 < \theta_t \leqslant 90°$, the director is aligned due to the Frederics effect perpendicular to the electric field, i.e. parallel to the cell surfaces, also except in the layers near the cell walls (Fig. 1c).

Given the liquid-crystal parameters, one can easily estimate the thickness of the wall layers in which the director has its initial alignment in a high-frequency electric field. Assuming that their thickness equals the electric coherence length ξ_E[8] we have

$$\frac{2\xi_E}{L} = \frac{2}{U_m}\sqrt{\frac{4\pi K_{33}}{|\Delta\varepsilon|}} = \frac{2}{\pi}\frac{U_c}{U_m},\tag{1}$$

where L is the thickness of the liquid-crystal layer, U_c is the threshold voltage of the Frederics effect, U_m is the voltage applied to the cell, and K_{33} is the elasticity coefficient. Substituting the threshold $U_c \approx 2.5$ V and operating voltages into Eq. (1), we find that at $U_m = 50$ V the wall layers occupy less than 3% of the liquid-crystal volume. Hence, if a sufficiently stong high-frequency field is applied to the cell, the resulting structure can be considered planar (with zero tilt angle, $\theta_t = 0$), and the error in the maximum phase difference Φ_{max} will be less than 3%. The error in Φ_{max} is maximum at large initial tilt angles, $\theta_t \to 90°$, because the real orientation of the director in the wall layer is not known.

If the polarization vector of light incident on the nematic liquid-crystal cell makes a certain angle $\phi_0 \neq 0, 90°$ with the
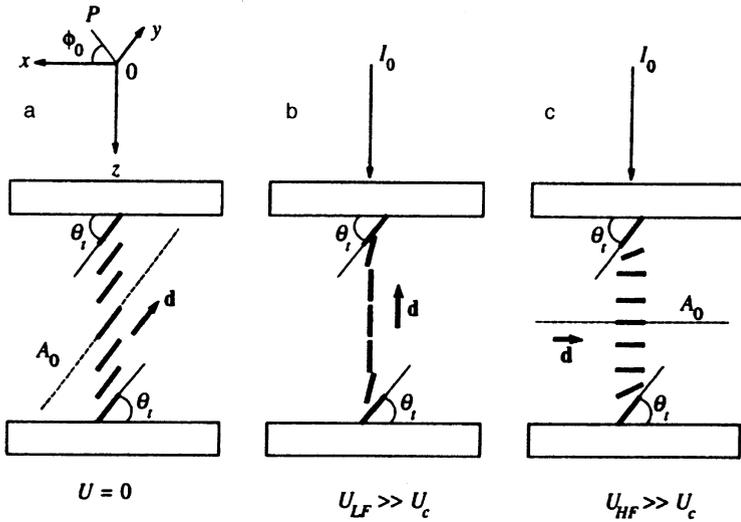
FIG. 1. Director orientation distribution in a liquid-crystal cell (a) with the pretilt angle in the initial state, (b) in a low-frequency and (c) high-frequency field: d) director of the liquid crystal; θ_t) pretilt angle of liquid-crystal molecules at the orienting surfaces of the cell; A_0) optical axis; U_c) threshold voltage of the Frederics effect; P) transmitted polarization axis of the polarizer; I_0) light propagation direction.

xz-plane in which the liquid-crystal director lies (Fig. 1a), the light transmitted through the cell will generally be elliptically polarized. The phase difference Φ between the ordinary and extraordinary components is uniquely related to the effective extraordinary refractive index $n_e(z)$ as a function of z due to the realignment of the director by the electric field. In monochromatic light at wavelength λ, the phase difference Φ is[9]

$$\Phi = \frac{2\pi}{\lambda} L\Delta n,$$

$$\Delta n = \frac{1}{L}\int_0^L [n_e(z) - n_o]dz,$$

(2)

and the effective extraordinary refractive index $n_e(z)$ is

$$n_e(z) = \frac{n_o n_e}{[n_o^2 \cos^2 \theta(z) + n_e^2 \sin^2 \theta(z)]^{1/2}},$$

(3)

where n_o and n_e are the ordinary and extraordinary refractive indices of the liquid crystal, and $\theta(z)$ is the angle between the x-axis and director at a point with coordinate z.

If the analyzer downstream of the cell has a transmitted light polarization vector perpendicular to that of the polarizer upstream of the cell, the change in the intensity I of transmitted light can be recorded as a function of the phase difference Φ:

$$I = I_0 \sin^2(2\phi_0)\sin^2(\Phi/2),$$

(4)

where I_0 is the intensity of the incident linearly polarized light.

The external electric field changes the distribution of the director over the cell thickness, $\theta(z)$, so it also changes the distributions of Δn and Φ. As the phase difference varies, the transmittance of the liquid-crystal cell between the crossed polarizers oscillates. The maximum amplitude of the light intensity oscillations corresponds to the angle $\phi_0 = 45°$, and the maximum number of oscillations recorded in a transition from the planar orientation with $\theta_t = 0$ to the homeotropic state is approximately $(n_e - n_o)L/\lambda$.

The phase difference in a liquid-crystal cell with the initial director alignment at angle θ_t (Fig. 1a) is

$$\Phi_t = \frac{2\pi L n_o}{\lambda}\left[\frac{n_e}{[n_e^2 - (n_e^2 - n_o^2)\cos^2 \theta_t]^{1/2}} - 1\right].$$

(5)

At $\theta_t = 0$, Eq. (5) yields an expression for the phase difference in a planar liquid crystal cell (maximum phase difference Φ_{max}):

$$\Phi_{max} = \frac{2\pi L n_o}{\lambda}\left(\frac{n_e}{n_o} - 1\right) = \frac{2\pi L}{\lambda}(n_e - n_o),$$

(6)

which equals the phase difference in a cell in the maximum high-frequency field (Fig. 1c). The phase difference Φ_t in a cell with unknown tilt angle can be measured experimentally by applying a low-frequency voltage to it. The final liquid-crystal orientation in this case is homeotropic (Fig. 1b). One can easily derive from Eqs. (5) and (6) an expression for the pretilt angle in terms of the phase difference Φ_t in the liquid-crystal cell at zero voltage and the maximum phase difference Φ_{max}:

$$\cos \theta_t = \frac{n_e}{\sqrt{n_e + n_o}}\frac{\sqrt{k(2n_o + k\Delta n)}}{n_o + k\Delta n},$$

(7)

where $k = \Phi_t/\Phi_{max}$, and $\Delta n = n_e - n_o$ is the optical anisotropy of the liquid crystal.

3. EXPERIMENTAL RESULTS

In our experiments, we used nematic liquid crystals in which the frequency of the dielectric anisotropy inversion at room temperature was $f_0 = 8$ kHz, the low- and high-frequency dielectric anisotropies were $\Delta\varepsilon_{LF} = 5.1$ ($f = 200$ Hz) and $\Delta\varepsilon_{HF} = -4.2$ ($f = 50$ kHz). Different pretilt angles of the director were realized by rubbing thin polyimide films[10] doped with surface-active substances, through light-induced optical anisotropy in polyimide films doped with dichroic dyes, and in polyvinylmethoxycinnamate films.[11]
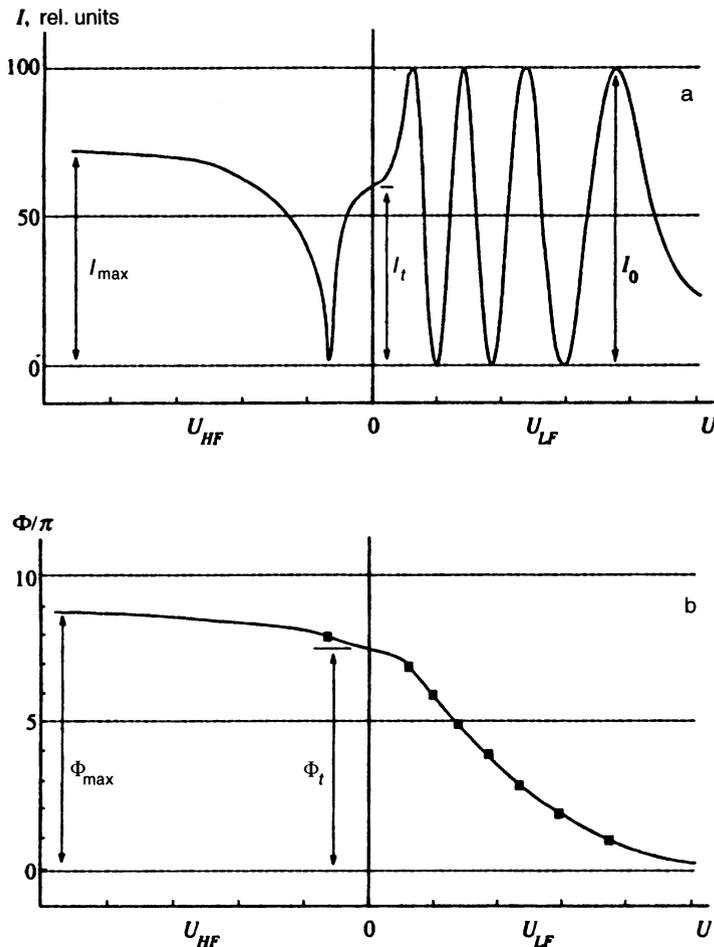
FIG. 2. (a) Typical oscillating curve of light intensity transmitted across a planar cell between two crossed polarizers and (b) optical phase difference as a function of voltage. Φ_t is the phase difference in the initial state ($\theta_t = \approx 20°$; is the Φ_{max}) phase difference in the planar configuration at the maximum high-frequency voltage (equivalent to $\theta_t = 0$); $n_e = 1.77$, $n_o = 1.54$, $\theta_t \approx 20°$, $L = 11.8 \mu m$, $\lambda = 632.8$ nm.

Liquid-crystal cells were manufactured from glass plates with conducting ITO layers; the liquid-crystal layer thickness was defined by 10–20-μm Teflon films.

Figure 2a shows a typical oscillating curve of the optical transmission of a liquid-crystal cell with an initial tilt angle $\theta_t = 20°$ placed between two polaroids with an angle $\phi_0 = 45°$ between them. The voltage scale is nonlinear for better demonstration of the curve at low voltages. The right side shows the signal versus low-frequency voltage and illustrates the crystal realignment to the homeotropic structure, the left side shows the signal versus high-frequency voltage and its realignment to the planar structure. The phase difference versus low- and high-frequency voltage derived from these measurements is given in Fig. 2b. Under a high low-frequency voltage ($U_{LF} > 50$ V), when the crystal alignment is homeotropic, the phase difference is approximately zero. Under a lower voltage, the director returns to its initial orientation and the phase difference increases. The initial phase difference Φ_t with the tilted director orientation is given by as the increment of the phase difference as the low-frequency voltage decreases from the maximum corresponding to the homeotropic structure to zero. The maximum (total) phase difference Φ_{max} corresponds to the planar orientation of the liquid crystal, and is the sum of the initial phase difference Φ_t and the added phase difference owing to the high-frequency voltage $U_{HF} \approx 50$ V. The phase differences Φ_t and Φ_{max} were derived from oscillating curves of the transmitted

light intensity versus voltage (Fig. 2a) using the formula

$$\Phi_i = \pi \left(N_i + \frac{1}{2} \right) + (-1)^{N_i} \left[2 \arcsin \sqrt{\frac{I_i}{I_0}} - \frac{\pi}{2} \right], \qquad (8)$$

where I_i is the transmitted light intensity at the point where the phase difference is determined, I_0 is the maximum transmitted light intensity, usually observed at the first transmission maximum corresponding to a phase difference of π, N_i is the number of extrema in the transmission curve between the first maximum and the point where the phase difference is determined.

Table 1 lists measurements of θ_t in liquid-crystal cells with different tilt angles. In order to compare and assess the applicability of the technique, measurements of the same cells using the rotating-crystal method are also given.

One can see in Table 1 that the agreement between the two techniques is the best in the range of tilt angles between 2.6 and 12.6°. It can be shown that the accuracy of the op-

TABLE I. Measurements of θ_t (deg).

Cell number	3	1	4	2	5	6	7
Phase-delay method	1.1	2.6	3.2	3.4	8.3	12.6	81.7
Crystal-rotation method	0.5	2.5	3.4	3.4	8.2	12.5	83.5

tical phase difference technique is highest at a tilt angle of about 45°, and deteriorates at the edges of the interval [0°,90°].

4. CONCLUSION

The proposed technique for measuring the tilt angle of liquid crystals with respect to orienting surfaces based on measurements of the phase difference at electric field frequencies corresponding to opposite signs of dielectric anisotropy does not require sophisticated experimental equipment or cumbersome calculations and has high accuracy over the range of tilt angles between ≈0 and ≈90°. Note that the technique is capable of measuring angles in the range 30–60°, which is not accessible to the rotating-crystal method. The limitation of our technique is that the crystal should have special dielectric properties, namely inversion of the dielectric anisotropy sign with frequency. The method may be useful in studying new orienting plates and developing techniques of surface processing. An important advantage of the method is the possibility of determining absolute values of initial tilt angles in liquid-crystal layers with different tilt angles on the two surfaces (after the appropriate modification of formulas), which is not possible when using either rotated-crystal or magnetic techniques.

[1] G. Baur, V. Wittwer, and D. W. Berreman, Phys. Lett. A **56**, 142 (1976).
[2] H. A. van Sprang, Mol. Cryst. Liq. Cryst. **199**, 19 (1991).
[3] K. Suzuki, K. Toriyama, and A. Fukuhara, Appl. Phys. Lett. **33**, 561 (1978).
[4] T. J. Scheffer and J. Nehring, J. Appl. Phys. **48**, 1783 (1977).
[5] N. O. Spaulding, W. E. Estes, V. E. Fainzilberg, and W. E. Hatfield, SID 90 DIGEST, 201 (1990).
[6] W. H. de Jeu, C. J. Gerritsma, P. van Zanten, and W. J. A. Goossens, Phys. Lett. A **39**, 355 (1972).
[7] M. I. Barnik, L. M. Blinov, A. V. Ivashchenko, and N. M. Shtykov, Kristallografiya **24**, 811 (1979) [Sov. Phys. Crystallogr. **24**, 463 (1979)].
[8] A. S. Sonin, *Introduction to the Physics of Liquid Crystals* [in Russian], Nauka, Moscow (1983).
[9] V. G. Chigrinov, Kristallografiya **27**, 404 (1982) [Sov. Phys. Crystallogr. **27**, 245 (1982)].
[10] M. I. Barnik, V. V. Lazarev, E. E. Maltzev, and N. M. Shtykov, submitted to Mol. Mat.
[11] A. G. Dyadyusha, V. M. Kozenkov, T. Ya. Marusii *et al.*, Ukr. Fiz. Zh. **36**, 1059 (1991).

Translation was provided by the Russian Editorial office.