Ellipsometric study of vertically aligned nematic liquid crystals

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The director tilt angle distribution in vertically aligned nematic liquid crystal displays has been investigated by means of variable angle spectroscopic ellipsometry. Liquid crystal vertical alignment has been realized by thermal evaporation of SiO_x. By changing the deposition angle, it is possible to control the pretilt angle. The director profile inside the sample was inferred by reflection and transmission ellipsometric measurements. The tilt angle distribution inside the cell versus the applied voltage is reported and eventually, comparing it with the simulations from the elastic theory, the anchoring energy has been obtained.
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Vertically aligned nematic (VAN) liquid crystal displays (LCDs) are currently the preferred option for consumer electronics devices such as large area flat TV displays, or high performance projection microdisplays. The angle between the LC director and the normal to the sample plane is called tilt. The tilt profile is ultimately determined by the external applied field and the pretilt, defined as the tilt angle on the surfaces at zero field. The control of pretilt and tilt angle distribution inside VAN cells is crucial for many applications,[1,2] because it affects the contrast and the response time. A number of methods to measure the pretilt angle have been proposed.[3] For display applications the control of the pretilt must be supplemented with the knowledge of the director distribution inside the pixel as a function of the applied voltage.[4] In this letter, we analyze the director tilt angle distributions inside the cell by means of variable angle spectroscopic ellipsometry measurements: thickness of alignment layers and LC layer, pretilt angles, anchoring energy, tilt profile, and optical constants are provided.

Cells were prepared with polished glass plates, pixels were imprinted on indium thin oxide (ITO) coating by photolithography. Vertical alignment was induced by evaporation of SiO_x onto the glasses. Different pretilt angles were obtained by changing the SiO_x evaporation angle. Coated substrates with antiparallel evaporation directions were assembled into sandwich cells with a gap of 4 μm filled with MLC6608 (Merck) nematic LC mixture with negative dielectric anisotropy, $\Delta\varepsilon=-4.2$. The LC physical properties are listed as follows: $K_{11}=16.7$ pN, $K_{33}=18.1$ pN, $\varepsilon_\parallel=3.6$, $\varepsilon_\perp=7.8$, $\gamma_1=0.186$ Pa, $n_o=1.475$, and $n_e=1.558$. All the data were measured at 25 °C and $\lambda=633$ nm. Applying a square form 1 kHz alternative voltage V, which exceeds the threshold value V_{th}, a nonlinear θ profile is induced along the cell gap, as shown in Fig. 1. Due to the presence of a pretilt, to speak of threshold voltage is not properly correct. However, for practical electro-optical characterization it is common to define an effective threshold voltage V_{th} as the voltage that corresponds to a 10% gray level. The z-axis corresponds to the normal to the cell plane, cell layers stay in xy plane; the zenithal angle θ (tilt) is the one between the LC director $\vec{n}$ and the z-axis; the azimuthal angle ϕ is the one between the y-axis and the director projection on the xy plane. The twist angle is defined as $\Delta\phi=\phi(z)-\phi(0)$. In this work nontwisted cells have been studied.

At oblique deposition the SiO_x layer has the sand dune like shape, which is responsible for *LC* alignment with the pretilt angle θ_o on a substrate. Cells with SiO_x deposition angles 50°, 67°, 70°, and 80° have been fabricated. Higher SiO_x deposition angles provide higher pretilts. The higher the pretilt is, the faster the display, but the lower the contrast. A good compromise is to use asymmetric cells with different angles of SiO_x deposition on either cell substrate. This case gives rise to a nonsymmetric director distribution with respect to the central plane of the cell.

The electro-optical response has been studied in transmissive mode by placing the cells between crossed polarizers. Obtained values for contrast ratio (transmittance ratio in on and off states), V_{th}, rise time t_R, and fall time t_F (defined as the time interval between 10% and 90% values of the maximum transmittance) are presented in Table I.

For the SiO_x deposition angle of 80° both response time and threshold voltage of a LC cell are reduced with respect to the case of 50° but the contrast ratio becomes unacceptably low. A good compromise has been found for the asymmetric cell "50-80:" a good contrast ratio is maintained and lower V_{th} and faster response are still provided.

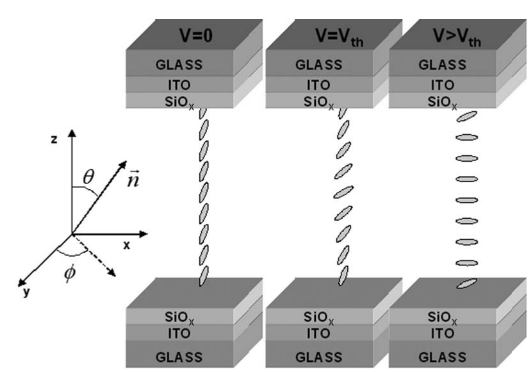

FIG. 1. Director tilt angle distribution in VANs for different voltages.

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TABLE I. Electro-optical parameters for cells with different angles of SiO_x deposition on the two substrates.

Deposition angles (deg) (top-bottom glass)	Contrast ratio	V_{th} (V)	t_R (ms)	t_F (ms)
50-50	110	2.75	5.0	5.0
50-80	80	1.75	3.6	5.2
80-80	10	1.00	0.5	6.8

To study the induced pretilt versus SiO_x evaporation angle, optical characterization of cells has been performed by the variable angle spectroscopic ellipsometer from J. A. Woollam Co., Inc. Ellipsometry is based on measurements of the relative phase change Δ and the relative amplitude change Ψ for two orthogonal polarizations of incident light after transmission (or reflection) by the sample. Obtaining quantitative information on the wanted parameters requires setting of a physical model of the sample and a fit procedure matching the data generated by that model with the experimental ones. In spite of the indirect measuring method of LC director orientation inside a cell, ellipsometry is very sensitive to it, but the accuracy of results strongly depends on the adequacy of the physical model, which has to take into account refractive indices and thickness of each layer of a multilayered cell.[5,6]

Let us first consider a set of measurements on the sample without external applied field. A three-step strategy was adopted for measuring pretilt, optical constants and thicknesses of the cell layers. In this case, the tilt distribution has to be either uniform or linear along normalized thickness d, thus being described by two fitted parameters A and B,

$$\vartheta(d) = A + Bd. \tag{1}$$

In the first step, spectroscopic reflection measurements were carried out from 400 to 1700 nm. Being more sensitive to refraction index variation, these data were used to obtain a first evaluation of the cell thickness and ordinary and extraordinary indices. Zero pretilt has been assumed at this stage. These measurements had been repeated for several incident angles of the impinging light to improve the accuracy of the results. More details on the use of ellipsometry to get accurate values of LC optical constants can be found in Ref. 6. In the second step, transmission ellipsometric measurements were performed at a fixed wavelength (633 nm) versus the incident angle from $-15°$ to $60°$. Being very sensitive to the sample birefringence, by the analysis of these data we obtained a first evaluation of the pretilt angles at the two surfaces, taking as fixed parameters the thicknesses and optical constants. In the third step, we analyzed reflection and transmission data together, keeping the previous values, obtained in steps 1 and 2, as starting point for a global fitting procedure that gave us the most accurate values for layer thicknesses, refractive indices, and pretilt at the top and bottom surfaces of the LC cell.

The measured pretilt values with a quoted error of 1°–2° are shown in Table II. A linear variation in the tilt angle occurs in asymmetric cells due to the different pretilts on the two substrates. A smaller variation is found in symmetric cells as well. This is attributed to imperfections in the repeatability of the deposition process. Measurements were performed from both sides of the cell in order to double check the reliability of results, showing in all cases the inverted pretilt values for inverted positions of the cell. Pretilt data for different samples, presented in Table II, are plotted in Fig. 2.

TABLE II. Pretilt in cells with different SiO_x deposition angles: ϑ_{top} is the pretilt at the upper alignment substrate; ϑ_{bottom} is the pretilt at the lower alignment substrate.

SiO_x deposition angles (deg)	ϑ_{top} (deg)	ϑ_{bottom} (deg)
50-50	0.9 ± 1	4.6 ± 1
67-67	4.2 ± 1	7.1 ± 1
70-70	7.4 ± 1	10.2 ± 1
80-80	26 ± 2	32 ± 2
50-70	0.0 ± 1	6.1 ± 1
50-80	1.2 ± 1	25.2 ± 1

Let us now discuss the case of the VAN cell distorted by an applied electric field. Transmission measurements (at $\lambda = 633$ nm) versus the incidence angle of the impinging light was carried out for voltages between 0 and 5 V. This choice was motivated by the high sensitivity of the measured data to the local optical axis orientation that coincides with the local director one. Once again the main problem in getting the values of the physical parameters in an inversion problem like this is to have an adequate physical model. Let us briefly recall that the ellipsometer software first takes into account the sample as a multilayered optical medium, with input parameters for each layer that can be fitted or not, and generates data for the Ψ and Δ curves. Then, generated curves are compared with the experimental ones and the procedure is iterated to minimize the mean square error.

For the case of LC, a widely accepted model for tilt distribution is based on the Frank theory of static deformation.[4,7] In general an anisotropic LC-to-wall interfacial energy per unit area can be expressed as a series of Legendre polynomials $P_{2n}(\cos\theta)$.[8] However for small changes in the boundary tilt angle with respect to pretilt, as in our case, one can retain only the first term $P_2(\cos\theta)$, which gives the Rapini–Papoular boundary condition formula.[9] Thus, we have implemented a software program able to calculate the director profile inside the cell for varying input parameters, like LC optical, dielectric, elastic constants, pretilt angles, driving voltage, and anchoring energy constants, including the possibility to feed different values at each surface to give account to asymmetric cells.

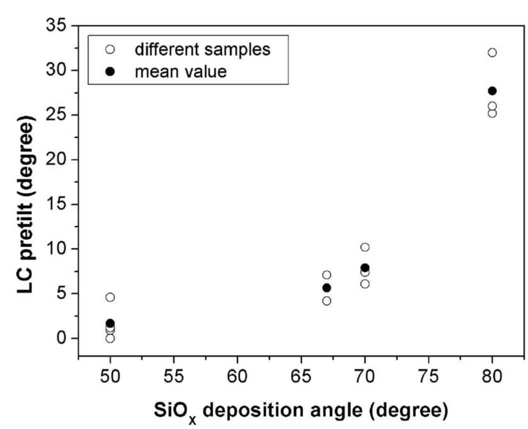

FIG. 2. Pretilt vs SiO_x deposition angle measured by ellipsometry. Mean values are shown as well to give pretilt dependence on SiO_x deposition angle.

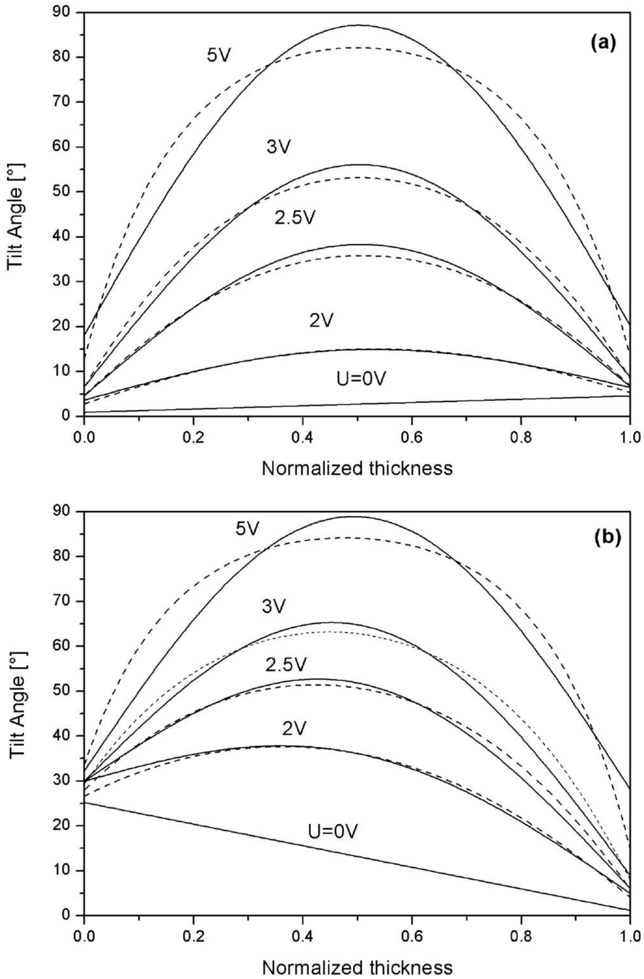

FIG. 3. Tilt profile simulated by the elastic theory (dashed lines) and by the sinusoidal model of the ellipsometry software (full lines) vs applied voltage: (a) symmetric cell 50-50 and (b) asymmetric cell 50-80.

It is worth noting that due to the closed structure of the ellipsometer software, we could not directly feed our simulation results into the fit procedure of the ellipsometer data. Thus, we made an off-line fit: using the simulated profile achieved from our software, we generated data curves of the ellipsometer parameters Ψ and Δ. These are then compared with the experimental ones in order to minimize the mean square error. During simulations the input values for all the relevant parameters, apart from the anchoring energy, were taken either from the literature (e.g., elastic and dielectric constants) or from our measurements at zero field (e.g., refractive indices and pretilt) and kept constant. The anchoring energy (allowing different values at both surfaces) was the only fitting parameter, letting the applied voltage as the external parameter, labeling the various tilt profiles, which are represented by dashed lines in Figs. 3(a) and 3(b). A global fit, considering all the curves at six different voltages (1.5, 2.0, 2.5, 3.0, 4.0, and 5.0 V), was performed in the case of the two cells 50-50 and 50-80, eventually getting for the anchoring energy W the value $(2.0 \pm 0.5) \times 10^{-4}$ J/m^2 for both cells and for both surfaces of each cell, in accordance with the previous values measured in the literature on similar interfaces.[4] It is worth noticing that the quoted error in the anchoring energy is relatively small as compared with other methods often providing only an order of magnitude. Moreover, the greater is the number of curves fitted at the same time, the more accurate is the obtained result.

In the general case, it is not possible to find an analytical solution for the steady state director reorientation at any applied voltage and for this reason the fitting section of the ellipsometer software could not be used and the above procedure was followed. However, for small voltages below 3 V, the simulated profile can be adequately approximated by a sinusoidal function, which is in accordance with an analytical solution of the Erickson–Leslie equation in the small angle approximation.[7,10] So, we introduced the sinusoidal term with amplitude C to Eq. (1), getting

$$\vartheta(d) = A + Bd + C \sin(\pi d). \tag{2}$$

The tilt distribution of Eq. (2) was used as a physical model in the ellipsometer software and the parameters A, B, and C were fitted, matching generated Ψ and Δ curves with experimental transmission data. The obtained tilt distributions are shown in Fig. 3 by full lines. A comparison between full and dashed lines in Fig. 3 from one hand confirms that our off-line fitting procedure is well acceptable because of the good matching among the numerically simulated and the analytical curves (2.0 and 2.5 V), and from the other hand shows that for higher voltages (3 and 5 V) the sinusoidal model fails to give an adequate representation of the actual director reorientation.

In conclusion, control and knowledge of tilt and azimuthal angle are critical for the performance of any VAN LCDs. Their distributions inside the LC cell can be controlled and designed using an appropriate alignment layer. VAN cells with different pretilts, depending on the angle of SiO_x alignment layer deposition, have been prepared and their parameters have been measured with spectroscopic ellipsometry. The tilt distribution inside a VAN cell has been obtained both in the case of undistorted and field oriented profile. A new way, based on ellipsometry, to measure the anchoring energy with a good accuracy has been proposed.

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