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# Molecular Crystals and Liquid Crystals

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### Molecular Orientation in Twisted Liquid Crystal cells

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## Molecular Orientation in Twisted Liquid Crystal Cells

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A useful model of the molecular orientation within twisted liquid crystal cells has been developed by Berreman (1). This model predicts a surprisingly large homogeneous component of orientation at fields at which a typical display is "on". It also predicts the retention of twist at voltages considerably above the normal "turn-on" voltage. At normal operating voltage the molecular configuration of partially homogeneous molecules with twist confined to the center of the cell can produce considerable retardation of off-axis polarized light. This degrades readability of the display.

A method of verifying this predicted molecular structure has been developed utilizing the interference fringes formed by reflections at the internal electrode-liquid crystal interfaces. Using HeNe laser light, these interference fringes are of considerable amplitude and permit an accurate measure of refractive index and birefringence. In 90° twisted cells, these interference fringes exhibit a 90° asymmetry. They easily show changes in molecular configuration at voltages well above the normal turn-on voltage.

The distribution of director tilt and turn angles within a 90° twist field effect LCD is of fundamental concern. Berreman has calculated this distribution for MBBA, using Oseen-Frank elastic theory. His calculations (normal incidence with parallel polars) show that essentially 100% transmittance can be achieved when there is a surprisingly large homogeneous component of molecular tilt. His model (Figure 4 of Reference 1) also indicates retention of twist at voltages twice that required to obtain essentially 100% transmittance. This paper is concerned with the experimental verification and application of this model. The mathematical problems have been simplified, and measurements are recorded here only for normal incidence.

The instrumentation used here has been described elsewhere.<sup>2</sup> It utilizes a lightly-focused polarized helium-neon laser. The liquid crystal cell used here consisted of two glass plates, each coated with SnO<sub>2</sub> electrodes, and a director-orienting layer of an oxide vacuum-deposited at a high angle of incidence.<sup>3</sup>

The plates were separated by 12.7  $\mu$ m thick plastic strips. The cell was held together with clamps so that the plates could be rotated relative to each other and provide control of the twist angle. An ester blend of liquid crystal material was used. Its dielectric anisotropy reversed sign at approximately 100 KHz. Accurate measurement of voltage applied to the cell and transmittance through the cell were facilitated using a digital voltmeter. Symmetric square wave fields of 45 Hz and 450 KHz were used.

Figure 1 showed the transmittance versus voltage (45 Hz) curve for the  $90^{\circ}$  twisted cell plotted on a semilog basis for parallel,  $\theta = 0^{\circ}$ , and crossed,  $\theta = 90^{\circ}$ , polars. Care was taken to place the front surface director in a plane normal to the incident plane of polarization. This method of graphing data has been shown superior to the conventional linear presentation.<sup>2</sup> Gooch and Tarry have developed a simple equation by which one might

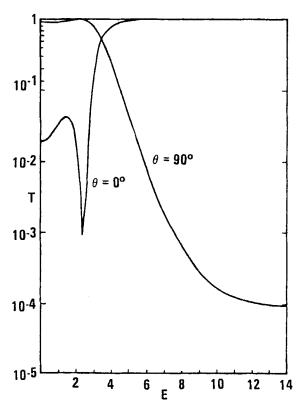


FIGURE 1 Log Transmittance vs voltage,  $90^{\circ}$  twist demountable cell filled with ester blend LC and separated by  $\frac{1}{2}$  mil spacers. Normal incidence, crossed ( $\theta = 90^{\circ}$ ) and parallel ( $\theta = 0^{\circ}$ ) polars. 45 Hz field.

hope to calculate retardation  $\Delta nd$  from transmittance T.<sup>4</sup> At normal incidence, the 90° twist cell should follow the equation

$$T = (\sin^2 \sqrt{1 + u^2})/(1 + u^2) \tag{1}$$

where

$$u = \pi d\Delta n/\lambda \tag{2}$$

These equations have been found to be accurate in predicting the variation of transmittance with cell thickness d when no voltage is applied to the cell. However, it has been found of little value in predicting transmittance when the molecules are under the influence of an electric field. This is most obvious in the 1-3 volt region of Figure 1 where two fringes (here called the residual retardation fringes) are expected on the basis of the known thickness d and birefringence  $\Delta n$ . Apparently, Eq. (1) is inappropriate when the director twist is not uniformly distributed throughout the Z dimension (normal to the cell surface). A better observation of these fringes is evident on the leading edge of the transmittance versus time recording when suddenly applying a field. Figure 2 shows recordings for 6 volt and 10 volt potentials. Two cycles of transmittance oscillation are seen in contrast to the single cycle of Figure 1. The amplitude of the fringes is seen to vary with voltage and does not follow Eq. (1). Presumably, molecules distribute tilt faster than turn when the field is suddenly applied. This leads to a change in retardation with uniform twist immediately after applying the field. The relative retardation fringes can be used to estimate the zero field retardation, hence to provide a measure of either birefringence or cell thickness, if one or the other of these quantities is known. Equation (1) also provides unrealistic values of retardation at voltages producing transmittances above 90% (parallel polars).

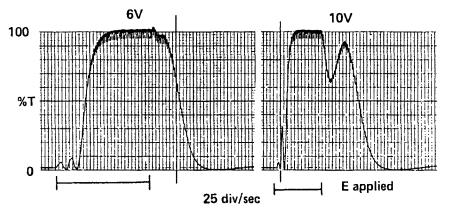


FIGURE 2 Transmittance vs time, same cell as Figure 1, 6 V and 10 V (45 Hz) potentials suddenly applied.

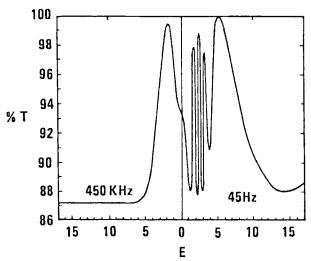


FIGURE 3 Transmittance vs voltage for 45 Hz and 450 KHz fields. Analyzer removed. Same cell as Figure 1.

Another potential method of determining molecular tilt involves measurement of the interference fringes arising from reflections within the cell. Figure 3 shows the variation of transmittance with voltage when the front surface director is in the incident plane of polarization and the analyzing polar is removed. In this case, the detector does not sense retardation per se, but responds to the optical path *nd*, according to the equation

$$T = (1 - R)^2 / (1 - 2R \cos \delta + R^2)$$
 (3)

where

$$\delta = 4\pi nd/2 \tag{4}$$

The reflectance R can be moderately large (0.034 in Figure 3) because of the high refractive index (approximately 2) of the  $SnO_2$  electrode layer. Of course, monochromatic light must be employed to obtain this interference, and the plate surfaces must be parallel within a fraction of a wavelength across the area of the beam. The cell plates used here were not perfectly flat, accounting for the nonuniform fringe amplitude.

Transmittance varies with voltage because the refractive index  $n_e$  in the plane of the E vector of the light beam varies with molecular tilt, hence with voltage. If the cell had been rotated 90°, placing the front surface director in a plane normal to the incident plane of polarization, a different interference pattern would have occurred (see Figure 3 of Ref. 2). At low voltages, the

light beam would have encountered only the ordinary index of the liquid crystal, and this ordinary index does not vary with molecular tilt.

Figure 3 includes data obtained with both low (45 Hz) and high (450 KH<sub>3</sub>) frequency fields. The sign of the dielectric anisotropy differs between these frequencies. The optic axis (director) aligns parallel to the low frequency field and normal to the high frequency field. At zero field, the director is tilted approximately 25° away from the plate surface, as dictated by the oxide layer on the plates.<sup>5</sup> As a consequence of this zero field tilt, there is no measurable threshold voltage. With the application of a low frequency field, the molecules move in the direction of homeotropy. High frequency fields orient the molecules more nearly homogeneous.

From the transmittance data of Figure 3, the product of refractive index change  $\Delta n$  and cell thickness d can be calculated as a function of voltage using Eqs. (3) and (4). A count must be kept of the cyclic repetition of  $\delta$ . The results are plotted in Figure 4. It should be noted that the  $\Delta nd$  plotted here is not identical with retardation. At no voltage does  $n_e$  approach 0, and it is impractical to reduce d to 0. Thus, the zero point for  $\Delta nd$  is purely arbitrary. Furthermore, the  $\Delta nd$  cannot be used to compute the true change in the extraordinary index at low frequency voltages above about 3 volts. In this regime, the E vector of the light beam does not follow molecular twist, but encounters the ordinary index of the system. At low frequency voltages below 3 volts and at all high frequency voltages, the light beam is predominantly influenced by the extraordinary index and in this regime, should show a voltage dependence determined almost exclusively by a  $\Delta nd$  equivalent to retardation.

The problem of accurately measuring the average extraordinary index,  $n_e$ , hence of the average director tilt, is simplified by studying cells of zero twist.

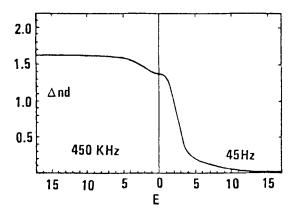


FIGURE 4 Change in optical path  $\Delta nd$ , vs voltage calculated from Figure 3.

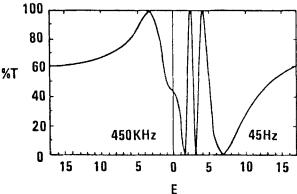


FIGURE 5 Transmittance vs voltage, parallel polars,  $\psi = 45^{\circ}$ , same cell as Figure 1 but with zero twist.

Figure 5 shows the variation of transmittance with voltage when a zero cell is oriented with the director rotated 45° from the plane of polarization. The analyzer was set parallel to the plane of polarization of the incident laser beam. Under these conditions, transmittance can be expected to oscillate between 0-100%, depending upon the retardation  $\Delta nd$ , hence upon the voltage applied. The same cell was used for Figure 5, as for the earlier figures, but in reassembling the cell after rotating the plates, the cell thickness did not return to the original value.

The equation governing the transmittance of light through a retarding element (45° orientation) between parallel polars at normal incidence is

$$T = 1 - \sin^2 \sigma/2 \tag{5}$$

where

$$\sigma = 2\pi d\Delta n/\lambda \tag{6}$$

These equations have been used to plot retardation  $\Delta nd$  as a function of voltage, and the results are graphed in Figure 6. In this case, there is no ambiguity in the transmittance at zero retardation. It has to be 1, and it is a virtual certainty that there will be less than one cycle of retardation between the highest voltage (low frequency) and the hypothetical input voltage at which the molecules are totally homeotropic and  $\Delta nd$  equals 0. At 16.8 volts (45 Hz), a retardation of 0.13  $\mu$ m is found. Clearly, all the molecules are not homeotropic.

There is less certainty concerning interpretation of the opposite end of the scale. The change in retardation on going from 16 volts at 45 Hz to 16 volts at 450 KHz is correctly graphed, but one cannot be sure that the molecules are totally homogeneous at 16 volts and 450 KHz. Of course, independent

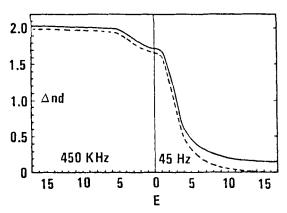


FIGURE 6 Change in retardation —— and change in optical path ---- vs voltage, zero twist cell, calculated from Figures 5 and 7.

methods for the measurement of cell thickness and material birefringence could be employed, and if sufficiently accurate, permit determination of the limiting retardation.

The reflection interference fringes obtained on rotating the zero twist cell to place the director in the plane of polarization of the incident beam can also be used to measure a  $\Delta nd$ . These fringes are seen in Figure 7. Since there is no cell twist, the light beam is always influenced by a component of the extraordinary index, and the data are useful at all voltages with the exception that transmittance at total homeotropy cannot be inferred. The total change

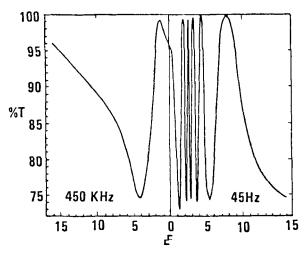


FIGURE 7 Transmittance vs voltage for the zero twist cell, analyzer removed.  $\psi = 0^{\circ}$ ,  $\theta = 0^{\circ}$ .

in  $\Delta nd$  on going from 16 volts at 45 Hz to 16 volts at 450 KHz is slightly larger from the interference fringe data than from the retardation data because of small differences in cell thickness when obtaining these two sets of data. The shapes of the two  $\Delta nd$  versus E curves are clearly similar. Superposition of Figure 6 on Figure 4 also shows a similar curve shape at voltages below three volts (45 Hz), indicating no gross effect of twist on  $\Delta nd$ .

The retardation data can be used to calculate the average molecular tilt using the equation

$$\bar{n}_e = \sqrt{n_0^2 + (n_e^2 - n_0^2)\cos^2 \alpha}$$
 (7)

where  $\alpha$  is the average angle between the director and the cell surface. The angle  $\alpha$  is plotted as a function of voltage in Figure 8. Of course, the director tilt will vary with the Z dimension of the cell at all voltages other than 0 volts. In preparing this graph, the retardation data from Figure 6 were used and the assumption was made that the retardation at  $\alpha = 0$  was 2.10. This leads to an uncertainty in  $\alpha$  that is inversely proportional to  $\alpha$ . For purposes of comparison, the linear transmittance versus voltage plot for the 90° twist cell is included in Figure 8. Of course, the tilt angle data apply to the untwisted cell while the transmittance data apply to the twisted cell, but the similarity of Figure 4 and 6 indicate relatively little interaction of tilt and turn. At 0 volts, the surface director tilt is seen to be 25° in agreement with other studies. At the  $E_{00}^{\parallel}$  point (4.3 volts), the average tilt is 63°. At the highest voltage used, the average tilt is still only 76°.

No attempt has been made during this study to measure the distribution of tilt angles as a function of the Z dimension. However, a comparison can be made between the average tilt angle obtained above and the values

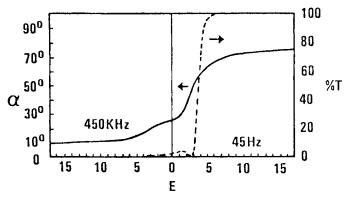


FIGURE 8 Average director tilt angle  $\alpha$  (zero twist cell) ———, and transmittance ---- for a 90° twist cell vs voltage.

calculated from Berreman's model. Because of the difference in surface director (zero field) tilt between the above cell and Berreman's hypothetical cell, the comparison can only be made at relatively high voltages. At the lowest voltage at which Berreman's cell transmits 100% ( $v/v_c = 2.56$ ), his average director tilt is  $63^\circ$ , while this study shows an average tilt of  $66^\circ$  when T equals 99%. At Berreman's highest voltage ( $v/v_c = 4.12$ ), his average tilt is  $73^\circ$ . At a comparable voltage, the average tilt observed is  $72^\circ$ , thus Berreman's distribution of twist is entirely compatible with the present observations.

A direct verification of Berreman's director turn model from normal incidence data is not easy experimentally. Intuitively, it is difficult to imagine a retention of twist when the central layer of molecules approaches as close to homeotropy as is indicated in Berreman's model. However, if the twist were to collapse, and all the molecules existed in the turn angles  $\beta = 0$  and  $90^{\circ}$ 

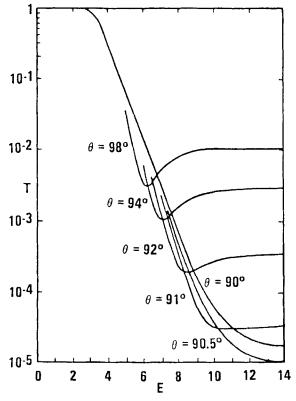


FIGURE 9 Log transmittance vs voltage (45 Hz) for a  $90^{\circ}$  twist cell at 6 angles  $\theta$  between the incident POP and the analyzer POP.

(Berreman's symbolism), the transmittance of a cell (normal incidence) between crossed polars would fall to 0, because no retardation can exist. Transmittance is clearly finite at all voltages below 14 volts in Figure 1. Nor have I ever seen a 90° twisted cell that exhibited 0 transmittance between crossed polars. However, the cell whose twist is confined to a narrow layer of highly tilted molecules will still partially rotate the plane of polarization of transmitted light. One might then expect to find some angle of the analyzer other than 90° (relative to the polarizer) at which a minimum transmittance is observed. Figure 9 shows log transmittance versus voltage curves for a 90° twist cell at several analyzer positions. The minimum is seen to become more pronounced and moved to lower voltages as the analyzer is rotated from its crossed ( $\theta = 90^{\circ}$ ) position. This is taken as an indication of twist retention at high voltage.

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