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Evaluation of optical anisotropy in the pretransitional regime in antiferroelectric liquid crystals

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By measuring the transmission through an antiferroelectric liquid crystal device (placed between crossed polarizers) as a function of both orientation and applied field in the so-called electroclinic-like, pretransitional regime the behaviour of the effective optic axis tilt angle (ψ) and effective optic anisotropy (Δn) is determined. The relatively poor alignment obtained in antiferroelectric liquid crystal devices is allowed for in the data interpretation and high quality results are presented.

1. Introduction

Antiferroelectric liquid crystals, AFLCs, [1, 2] are very promising materials for display applications. Their intrinsic non-linearity allows for a large number of rows to be passively multiplexed while a symmetrical driving scheme [3] ensures good resistance to ionic memory effects and makes grey scale feasible. However the process of switching is not fully understood and the achievable quality of alignment is not yet satisfactory for high quality displays. Two main factors can be identified as limiting the exploitation of AFLCs: (i) lack of local uniformity in alignment and (ii) the so-called electroclinic-like, pretransitional regime, which is observed for low fields. They both affect the achievable contrast and hence prevent the reproduction of a large number of grey levels. The former reduces the quality of black in the AFLC ground state, while the latter, characterized by a fast response [4], reduces the immunity to cross-talk data in passively addressed displays. The nature of this regime is still not understood fully, although there is strong evidence that this is not a pure soft mode electroclinic behaviour [5–7]. As the applied voltage is increased beyond the electroclinic-like pretransitional regime, switching follows, without any saturation of the optic axis rotation being observed.

Here we discuss a careful experimental investigation of this regime by means of accurate measurements of optical transmission of a device placed between crossed polarizers. Changes in optical anisotropy and of optic axis orientation have been separated as a function of applied voltage and are presented here, together with their mutual relationship.

Previous studies by others have been either insufficiently sensitive to changes in orientation and Δn to study the pretransitional regime in detail, or have not obtained conclusive results. For example the use of interferometric techniques by Oka *et al.* [8] allowed them to determine changes in orientation and refractive indices in AFLC switching, but only for full switching and not in the pretransitional regime under quasi-static conditions. Use of a random alignment allowed Buivydas *et al.* [7] to separate the effects of Δn changes from reorientation changes, but they do not present their functional form with voltage.

2. Modelling AFLC optics

Here we use simple low pre-tilt antiparallel rubbed polyimide aligned cells of thickness $\approx 3\mu\text{m}$, filled with the AFLC mixture Chisso CS4000 in the isotropic phase and allowed to cool slowly to room temperature. Observation of such cells under a polarizing microscope shows that a general feature is their strong tendency to form poor quality alignment with different extinction position for two types of domains being observed (see figure 1) this reduces the quality of the ‘black’ in the AFLC ground state. In carefully cooled cells, this leads to a ‘black’ state with about 2% of the maximum transmission, limiting the achievable contrast to less than 50:1.

Inside the domains, a spread in alignment orientation also exists, indicating that the structure in the cell is quite complex. It may however be reasonable to associate the two domain types observed with two dominant smectic layer orientations and this helps the data interpretation, being the key to the approach we take below. Further it should be noted that the relative density of

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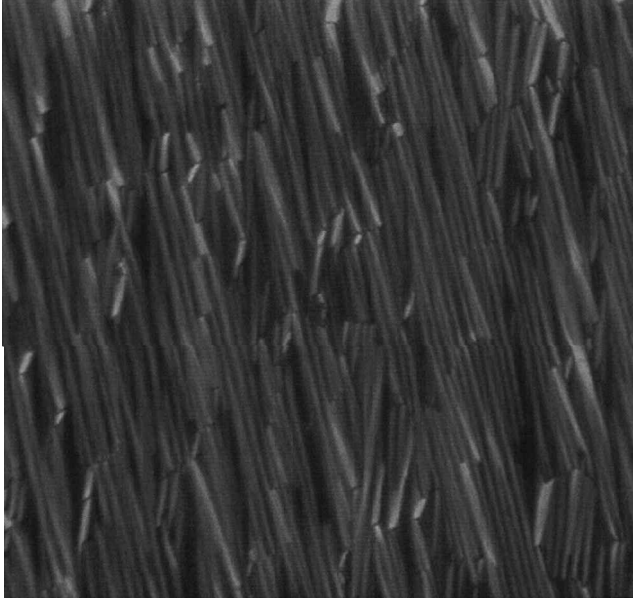


Figure 1. An image of the relatively poor alignment in an antiferroelectric liquid crystal.

the two types of domains varies considerably over the area of the cell—a further point which must be taken into account when considering the generation of several intermediate gradations of grey over a large area display.

This means that the usual expression for the optical transmission of a birefringent slab (placed between crossed polarizers) or optical anisotropy Δn and thickness d , whose optic axis makes an angle ψ with the direction of the incoming polarized light of wavelength λ , expressed by:

$$I = I_0 \sin^2(2\psi) \sin^2\left(\frac{\pi d \Delta n}{\lambda}\right) \quad (1)$$

where I_0 is the incident intensity on the slab, *cannot be used any longer*.

A more appropriate analytical description should take into account the real structure of the cell. We note that this can be done by splitting the contribution to the overall transmitted light intensity into those for the two types of domains, giving:

$$I = I_0 \{A_1 \sin^2(2\psi) + A_2 \sin^2[2(\psi - \Delta\psi)]\} \sin^2\left(\frac{\pi d \Delta n}{\lambda}\right) \quad (2)$$

where A_1 and A_2 describe the relative areas of the two parts of the domain structure, and $\Delta\psi$ their average orientation difference. In this expression ψ is therefore the angle which the optic axis of the domains weighted with area A_1 makes with the polarizer axis and $\psi - \Delta\psi$

is the angle of the optic axis for the domains weighted with area A_2 .

The existence of a spread in the orientation of the domains visible in figure 1 can now be neglected and the distribution of the orientation in each domain is approximated with the mean value. This is acceptable, because the spread within each domain type, of the order of a few degrees, is much smaller than $\Delta\psi$ and in practice it can be kept low if cooling into the AFLC phase is performed extremely slowly. Assuming the same optical anisotropy for both domains is also reasonable, as they appear to have the same structural form and between them the liquid crystal director appears rotated only in the plane of the cell. Further, no reconfiguration of the overall domain structure is observed for any field; the domains have been somehow frozen in after the phase transition and are not disturbed even during full switching.

This indicates clearly that the domain structure observed is due to the smectic layer structure in the cell, and not (for example) to surface switched states of the molecular director. As noted above this helps with the data interpretation because *we can treat director reorientation in the two types of domains independently if we assume the smectic layer structure remains fixed*.

A direct evaluation of $\Delta\psi$ can be made by independently measuring under the microscope the average orientation difference of the extinction positions of the two sets of domains. For our devices it is found to be $\approx 17^\circ$. Thus equation (2) with $\Delta\psi = 17^\circ$ allows a reasonable representation of the transmission of the AFLC device as a function of orientation angle (ψ) and optical anisotropy (Δn). It will be used in our analysis.

3. Experimental

Data are taken for a typical device as absolute transmission between crossed polarizers (i.e. normalized to unity for parallel polarizers) at a fixed wavelength of $\lambda = 632.8$ nm. A hot stage is used to maintain the device temperature at 30°C .

We are interested in determining the independent variations of ψ and Δn with voltage, so when a voltage is applied, contributions to transmission variations from changes of optical anisotropy and of optic axis direction must be separated. This can be done by mapping the intensity of the light transmitted by the cell as a function of orientation between crossed polarizers for different applied voltages. These voltages are applied as quasi-d.c. signals of 0 V, ± 2 V, ± 5 V, ± 10 V, ± 15 V. Measured data are shown as the discrete points in figure 2. Now the data are fitted by a curve given by:

$$I = A'_1 \sin^2[2(\psi - \phi)] + A'_2 \sin^2[2(\psi - \phi - 17^\circ)]. \quad (3)$$

ϕ , where ϕ is the offset in the optic axis orientation (which will vary with voltage due to the pretransitional, electroclinic-like, effect), and A'_1 and A'_2 , are the transmissions of the domains which will vary with voltage if any change in optical anisotropy (Δn) accompanies the effect.

As clearly seen from the plots, the agreement between experimental data and their fits is excellent. This means that neglecting the small spread does not appear to limit the accuracy of the extracted information for the field-induced changes in the optic axis. The best-fit parameter ϕ in equation (3) gives now a direct measurement of the optic axis rotation for an applied voltage by plotting $\phi(V) - \phi(V=0)$, shown in figure 3, where the evaluated maximum transmission and the main contribution A'_1 are also given. It is clear from figure 3 that the behaviour of director orientation with voltage is superlinear. This is in contrast with the common smectic A electroclinic effect where the re-orientation angle saturates at higher voltages for temperatures near the phase transition, and is always sublinear (or at most linear) [9]. Such direct evidence that the AFLC electroclinic-like, pretransitional behaviour is not a soft mode response will be important in developing theoretical understanding of the effect.

Knowledge of device thickness and light wavelength allows the optical anisotropy to be extracted from the

(a)

(b)

Figure 2. (a) Normalized transmission between crossed polarizers as a function of orientation for various applied voltages in the pretransitional electroclinic-like regime. (b) A highly expanded view around the maximum transmission. Experimental data shown with (+) for positive and (\times) for negative voltages; continuous lines represent best-fits using equation (3).

This is of course a variant of the form of equation (2), where, because of the normalization we are able to set I_0 to unity. The fitting parameters are then A'_1 , A'_2 , and

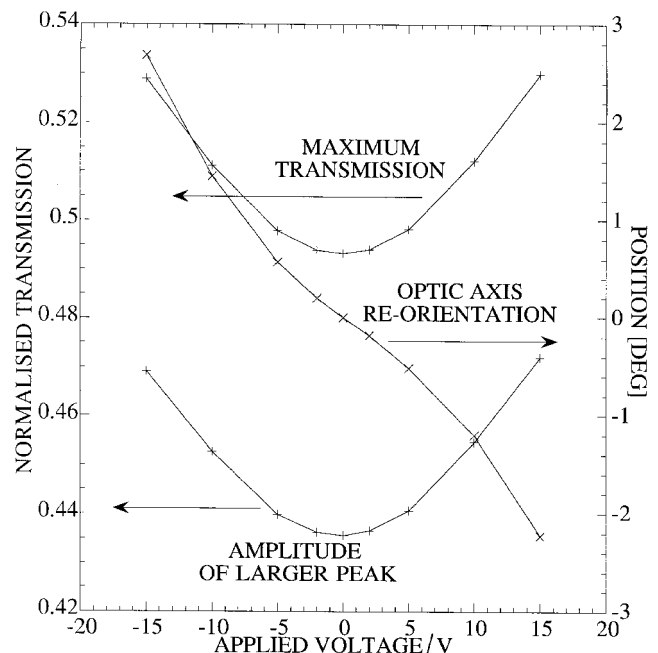


Figure 3. Maximum transmission between crossed polarizers as a function of voltage. The main amplitude coefficient and optic axis orientation are also shown.

transmission values evaluated from the best-fit functions by inverting

$$I_{MAX} = I_0 \sin^2 \left(\frac{\pi d \Delta n}{\lambda} \right). \quad (4)$$

While inverting the formula, the problem of multiple determinations for Δn could arise. This is however solved by recalling that the maximum transmission of the cell between crossed polarizers when the material is forced into either of the field-induced FLC states allows determination of $\Delta n = n_e - n_0$ for the aligned molecular director. Doing this (which requires > 35 V to be applied to our device) we obtain a value which is close to the maximum transmission between parallel polarizers; hence the $\lambda/2$ condition (which would correspond to $\Delta n = 0.11$) is approximately reached in the test cell. It should be noted that the next possible Δn value leading to peak transmission of unity in the FLC state would be $\Delta n = 0.32$, which is not reasonable.

The optical anisotropy of the herringbone AFLC ground state can be calculated by considering that its two refractive indexes are given by:

$$\begin{aligned} n_1 &= \sqrt{n_e^2 \cos^2 \theta + n_0^2 \sin^2 \theta} \\ n_2 &= \sqrt{n_e^2 \sin^2 \theta + n_0^2 \cos^2 \theta} \end{aligned} \quad (5)$$

where n_e and n_0 are the refractive indices in the FLC field-induced state. A typical value for n_0 is chosen and the cone angle value is used for θ , which we found equal (within the experimental error) to the value of 27.1° (at 25°C) claimed by the AFLC manufacturer. Then we can estimate $\Delta n = n_1 - n_2$ in the relaxed state. This is close to that for the first value on inverting equation (4), and breaks any degeneracy. The resulting behaviour of $\Delta n(V)$ is shown in figure 4.

It is clear that the behaviour of the optical anisotropy as a function of voltage is also superlinear. This seems to be further evidence that the process is different from the electroclinic behaviour experienced in the SmA phase where linear behaviour followed by a saturation at higher voltages can be observed for the optical anisotropy also [10]. It should be pointed out again that voltages used are still far from switching, and that no 'striped' domains, running along the smectic planes, are observed even after the d.c. bias has been applied for several tens of seconds.

From the above data it is also possible to extract the optical anisotropy versus the optic axis reorientations as shown in figure 5. It is worth noting that this is subtly different from the behaviour of Δn with voltage, being

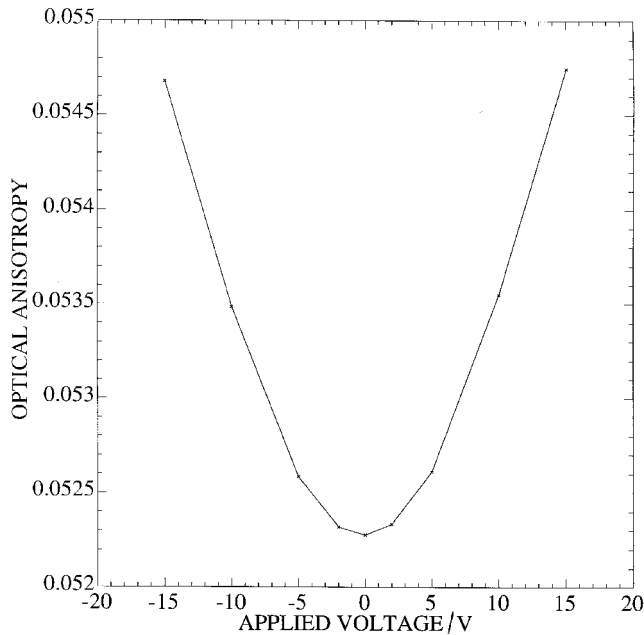


Figure 4. Optical anisotropy as a function of applied voltage.

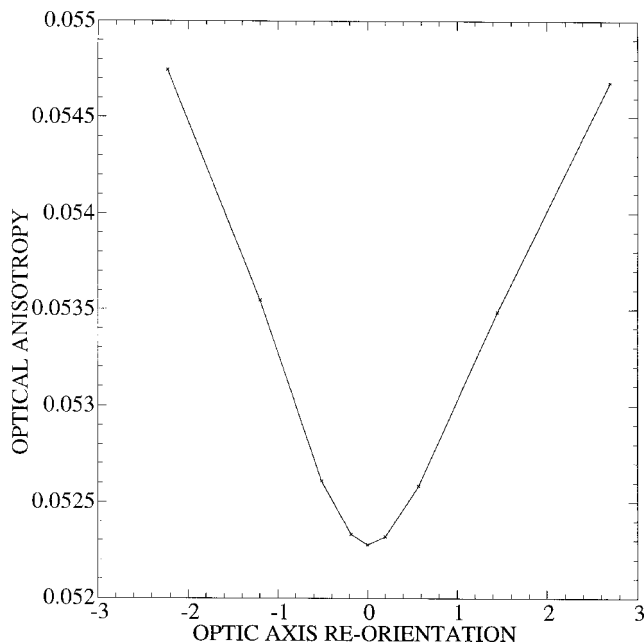


Figure 5. Optical anisotropy as a function of reorientation angle.

sharper around the zero reorientation region, and being almost linear for reorientation $> \approx 0.5^\circ$. This is interesting because it leads to information about the nature of the reorientation under an applied field, and how this is coupled to changes in the effective optical anisotropy.

These are important issues in identifying the mechanism of the pretransitional electroclinic-like effect, and we hope to discuss them further in a future publication.

4. Conclusions

We have presented a novel technique to evaluate accurately from the simple measurement of the optical transmission for optical anisotropy and the optic axis reorientation under an applied electric field in an AFLC showing the pretransitional behaviour. Disturbing effects of poor alignment have been taken into account in our analysis and thus their effect removed. Our procedure has been applied to the investigation of the so-called electroclinic-like, pretransitional regime, where we have found that the optic axis rotation does not show a linear dependence on the applied voltage even for very low voltages. It has also allowed us to extract the variations of the optical anisotropy with the applied voltage which we have shown to be superlinear as well.

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