Director-polarization reorientation via solitary waves in ferroelectric liquid crystals

I. Abdulhalim,[a] G. Moddel,[a] and N. A. Clark[b]
Center for Optoelectronic Computing Systems, University of Colorado, Boulder, Colorado 80309-0525

(Received 18 July 1991; accepted for publication 11 November 1991)

The spatial nonuniformity of the director field in ferroelectric liquid crystals during switching is demonstrated using the electro-optic response to a bipolar field and explained by director solitary wave motion.

Solitons appear liquid crystals (LCs) as a result of the nonlinear behavior of the director motion under external fields generally governed by differential equations of the reaction-diffusion type. Until now, solitons have been observed in nematics,[1,2] and possible soliton switching in ferroelectric liquid crystals (FLCs) in their helicoidal geometry has been demonstrated both experimentally[3] and theoretically.[4,5] In this letter, we report on a novel electro-optic effect in surface stabilized FLCs (SSFLCs), wherein the FLC is the dielectric in a transparent parallel plate capacitor thin enough for surface interaction to produce helix-free director states.[6] In particular, we show that during switching at high enough fields the director nonuniformity stabilized by the surface interactions evolves as two domain walls forming a kink-antikink pair propagating into the LC from the bounding plates via solitary wave motion of the sine-Gordon type.

In FLCs, the molecules are arranged in layers with the average orientation $\hat{\mathbf{n}}$ of the molecular long axis tilted at an angle θ from the layer normal $\hat{\mathbf{z}}$. The ferroelectric polarization $\mathbf{P}$ is locally normal to the $\hat{\mathbf{n}}$-$\hat{\mathbf{z}}$ plane and has an azimuthal orientation about $\hat{\mathbf{z}}$ given by the angle ϕ. Using particular alignment treatments of the surfaces, SSFLC cells can be obtained having planar smectic layers nearly normal to the plates and two different values of ϕ, (ϕ_1,ϕ_2) on the plates resulting in a splay deformation of the coupled $\mathbf{n}$-$\mathbf{P}$ distribution.[6] That is $\hat{\mathbf{n}}$ rotates over the tilt cone by an angle $(\phi_2-\phi_1)$ when progressing from one bounding plate to the other. The dynamic equation governing the motion of the dipoles in such a structure under an alternating (square wave) electric field, $\mathbf{E}=\pm|E|\hat{\mathbf{x}}$, applied parallel to the layers is given by the double overdamped sine-Gordon equation:

$$\eta_\phi \frac{\partial \phi}{\partial t}=K_s \frac{\partial^2 \phi}{\partial x^2}\pm |P|\,|E| \cos \delta \sin \phi + \left(\frac{\Delta\epsilon E^2 \sin^2\theta \cos^2\delta}{4\pi}\right)\cos\phi\ (\sin\phi - \sin\phi_0). \quad (1)$$

Here x is the coordinate normal to the cell plates, η_ϕ is the rotational viscosity associated with ϕ-motion, K_s is the splay elastic constant, $\Delta\epsilon$ is the dielectric anisotropy, θ is the molecular tilt angle, and δ is the layer tilt angle. If the director is parallel to the surface, the boundary orientations are ϕ_1 and $\phi_2=\pi-\phi_1$ where $\sin\phi_1=\tan\delta/\tan\theta$. Numerical solutions to Eq. (1) with fixed boundary conditions yield nonuniform structures during switching, including propagation of a single kink from one plate to the other.[5]

We report experiments on 10-μm-thick SSFLC cells using the FLC material Chisso 1014 between two optically flat substrates coated with transparent indium tin oxide electrodes of thickness 700 Å. The alignment of the FLC was achieved by the polymer rubbing technique with parallel rubbing directions on the two substrates. The aligning polymer was nylon 6/6. This treatment produced zig-zag free smectic C texture having the planar tilted layer structure. The $\mathbf{n}$-$\mathbf{P}$ configuration in absence of applied electric field had the $\mathbf{P}$ field splayed.[6] In order to probe the nonuniform structure during the switching, a symmetric square wave was applied to the cell [Fig. 1(a)]. The coupling of $\mathbf{E}$ to $\mathbf{P}$ produces, after a switching transient, a spatially uniform $\mathbf{n}$-$\mathbf{P}$ distribution with $\mathbf{P}=\pm|P|\hat{x}$ on the $\pm$ square wave cycle. The corresponding $\mathbf{n}$ orientations are indicated by the heavy lines in Figs. 1(a) and 1 (b). With normally incident light, the cell is oriented between crossed polarizers so that the transmittance during the positive and negative field intervals has the same level, i.e., with the optical polarization direction along Σ, the projection of the layer normal on the cell plates [Figs. 1(c) and 1(d)]. The evolution of the transmittance in time between these two levels is a probe of the transient structure during the switching. In this case, if $\hat{\mathbf{n}}$ is uniform during switching (free boundary conditions), extinction would be observed when $\hat{\mathbf{n}}$ reaches its midpoint on the cone, without dependence on the wavelength of the incident light λ. That is a minimum in the transmittance which would be observed generally at the midpoint of the switching. Our experimental results show (cf. Fig. 1) that this is not generally the case. Rather, depending on λ, a peak can appear rather than a dip.

Specifically, when the wavelength is such that the cell with the voltage applied is a multiple full-wave plate (MFWP) [Fig. 1(d)], the transmittance levels during the positive (+) and negative (−) field intervals are close to zero, but switching produces a single maximum in the transmission, with no evidence of the midpoint extinction expected for a uniform $\mathbf{n}$-$\mathbf{P}$. The switching transient exhibits the following general behavior. For the MFWP condition, the peak height and shape do not depend on the orientation of the cell with respect to the incident light

[a]Also with the Department of Electrical and Computer Engineering.
[b]Also with the Department of Physics.

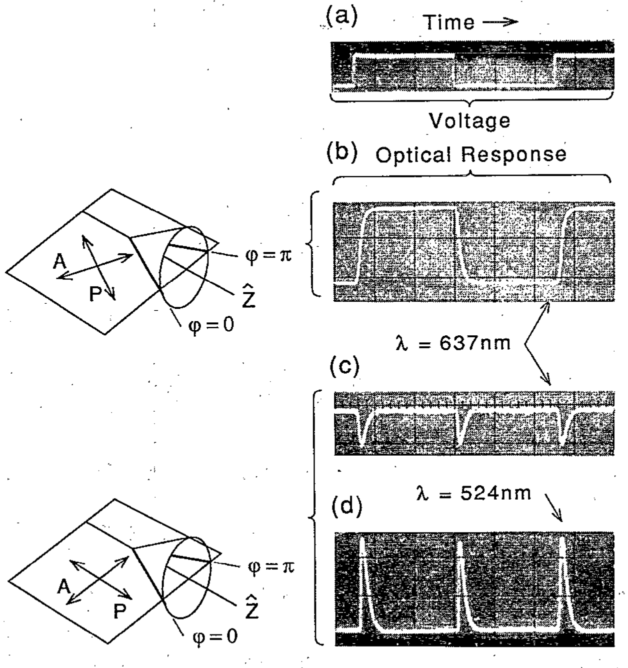

FIG. 1. (a) The bipolar square wave voltage applied to a 10-μm-thick CS-1014 cell (period = 5 ms, voltage = $\pm$50 V). (b$\rightarrow$d) resulting optical responses for the cell between crossed polarizers. (b) The response when the transmittance during the positive (negative) interval with the wavelength matching the multiple half wave plate (MHWP) condition. (c) The response with the polarizer oriented parallel to the layer normal with the MHWP condition $\lambda = 637$ nm so that the transmittance level is the same during the positive and the negative intervals. (d) As in (c), with the wavelength matching the M-Full-WP condition $\lambda = 524$ nm.

polarization. The larger the deviation from the MFWP condition, the larger the dependence on the cell orientation. When λ matches the multiple half-wave plate (MHWP) condition [Figs. 1(b) and 1(c)] the transmittance during the (+) and ($-$) intervals is close to 50% and a dip is observed. However, contrary to the case of uniform director switching where complete extinction occurs at the dip, the minimum transmission observed is generally nonzero. For small fields, a dip might appear at any wavelength and the asymmetric response is more pronounced. For intermediate fields, more than one peak (or dip) might appear depending on the wavelength and the cell thickness. As the field increases, the previous pattern develops generally towards one peak with increasing height till reaching a maximum at a certain field, then decreases again and saturates.

The behavior shown in Fig. 1 was observed on cells with different thicknesses using a variety of LC mixtures with various rubbed polymers and obliquely evaporated aligning layers. Although small variations do exist between different cells, such as the asymmetry in time of the response peak or the appearance of more than one peak, the general behavior is as presented above. We believe that these small differences are mainly a result of varying surface anchoring conditions and layer structure in different cells. For now, we wish to explain the results shown in Fig. 1 since this is the most commonly observed behavior. We present results on a 10 μm cell because such thick cells represent the more general case, as will be clarified below.

The measured cell transmission will be compared with the transmission obtained from the numerical solution of Eq. (1) to find the director distribution $\phi(r, t)$, followed by the application of the 4×4 matrix method[7,8] to find cell transmission versus time at a fixed wavelength. Numerical integration of Eq. (1) was performed using Gear's finite difference technique[9] with 1000 mesh points, different fixed boundary conditions ϕ_1, and $\pi - \phi_1$, and different initial profiles $\phi(z, 0)$. A negative step potential was applied first for a time interval $t- = 20$ ms and we calculated the profile $\phi(z, t-)$ at the end of this interval. This profile represents the ($-$) state. Then the field sign was reversed instantaneously and the profile $\phi(z, t-)$ was taken as the initial data for the calculated profiles $\phi(z, t)$ during the switching.[8]

The two possible initial solutions to Eq. (1) are either a uniform state or a linearly splayed structure. The initially splayed structure was chosen for the modeling because when the cells were observed under the microscope between crossed polarizers they did not exhibit complete extinction of white light. The splayed structure is represented by the profile $\phi = q_s z + \phi_1$ with splay wave vector $q_s = (\pi - 2\phi_1)/d$ so that $\phi_2 = \pi - \phi_1$. The simplest situation is when $\phi_1 = 0$. In this case, applying a negative field distorts the linear profile so that the state $\phi = 0$ is preferable. Above a certain field, all the cells switch to this state except for a thin localized region near the second boundary which can be described by the static sine-Gordon kink: $\phi \propto \arctan\{\exp[(z-d)/\Delta]\}$, with the width $\Delta \propto 1/(|E|)^{1/2}$. This profile is shown in Fig. 2(a) and labeled $t=0$. Reversing the field in this situation causes the above kink to propagate in the negative z direction with constant velocity $v \propto |E|^{1/2}$ and shape represented by the above solution[5] with z replaced by $z+vt$. The temporal evolution of the transmittance which corresponds to Fig. 2(a) is shown in Fig. 2(d). The transmittance pattern in this case consists of three identical peaks. We checked that this three-peak pattern remains the same as the field increases, with the peaks becoming narrower in time. The appearance of such a pattern of three identical peaks can be predicted easily without the numerical calculation. For this purpose, the kink, at any time, can be considered as a very thin region which is not seen by the light when its width becomes less than the wavelength. At any time during the time during the switching, the cell then behaves as a pair of birefringent slabs, one having the initial orientation and of thickness $d-vt$, and the other having the final orientation and thickness vt. The peaks in the numerical solution are birefringence interference fringes of fixed wavelength light passing through the two slabs as t increases. While the boundary condition $\phi_1 = 0$ exhibits the correct qualitative behavior, in which the transmittance during switching does not consist simply of a single dip, it does not yield a quantitative explanation of the experiment with regard to the number of peaks and the field dependence.

In an attempt to explain the experimental observations quantitatively, we modified the simulations. The only case which explains the experiments nicely is the initially

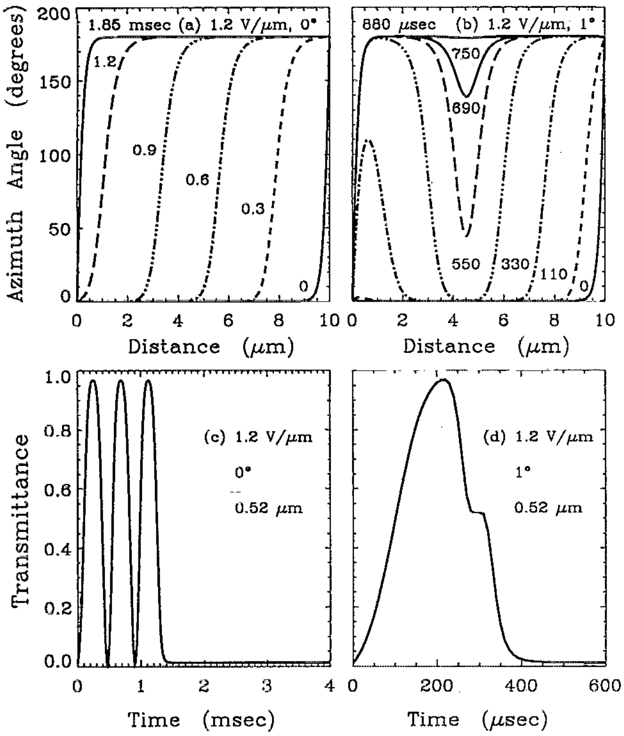

FIG. 2. Calculated azimuth angle profiles at different times [(a) and (b)] and the corresponding transmittance evolution [(c) and (d)] after the field sign was reversed. The profile at zero time was calculated after applying a negative step pulse for 20 ms to an initially linearly splayed cell with the fixed boundary orientations: (a) $\phi_1 = 0$ and $\phi_2 = \pi$, and (b) $\phi_1 = 1°$ and $\phi_2 = 179°$. The calculations performed using the parameters close to those of CS-1014: $P = 10$ nf/cm^2, $\theta = \pi/8$, $\Delta\epsilon = -1$, $\eta_\phi = 0.016$ p, $K_t = 10^{-7}$ erg/cm. The refractive indices were taken as $n_\perp = 1.5$ and $n_\parallel = 1.66$.

splayed structure with ϕ_1 in the range: $(0.01° < \phi_1 < 5°)$. For $\phi_1 > 5°$, more than one peak appears, and for $\phi_1 < 0.01°$, the pattern resembles the $\phi_1 = 0$ case. In Figs. 2(b) and 2(d), both the dynamics and the evolution of the transmittance are shown for the case of $\phi_1 = 1°$. The small field regime is not shown and it is not sensitive to such a small change of ϕ_1. In the high field regime and at the end of the $t- = 20$ ms interval, most of the cell switches to the $\phi_1 = 0$ state except for two narrow regions localized near the two boundaries where the maximum values of ϕ are the fixed values $\phi_1 = 1°$ and $\phi_2 = 179°$. When the field sign is reversed, the dipoles with higher ϕ, i.e., near the surfaces, start to switch first and kinks evolve from both surfaces. As the field increases, their speed becomes larger and they become narrower. The transmittance evolution exhibits a single peak which has a maximum height at a certain field. Then it decreases and saturates. In Fig. 3, we show a comparison of the measured and the calculated peak height normalized to the maximum value it attains versus $|E|$. The agreement between the measured and the calculated quantities is good.

Hence, a very small deviation of the fixed orientation of the molecules on the boundaries from the two states $\phi = 0$ and $\phi = \pi$, alters the electro-optic response drastically. This result is of particular interest because it demonstrates the sensitivity of the response of FLCs to the boundary conditions. Here, we should mention that the nature of the splayed structure is not crucial. Because the ϕ_1 which emerges is so small (in the range of 0.02° to 2°), it can be considered simply as a small deviation of the molecular director upward from the substrate plane, perhaps spatially nonuniform and not necessarily as a result of layer pretilt.

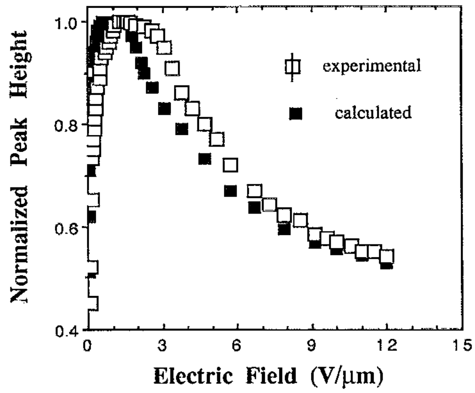

FIG. 3. Comparison between measured and calculated transmittance peak height normalized to its maximum value attained vs $|E|$. The experimental points are from measurements similar to Fig. 1 and the calculated ones are similar to those of Fig. 2(d).

In summary, we have explored the nonuniform director structure of FLCs during the switching by showing that at high fields, the transient intensity through crossed polarizers consists of a single peak rather than the dip expected for a spatially uniform director. This behavior was found to be explained by the creation of sine-Gordon kink-anitkink pairs which propagate in the FLC until they annihilate each other. The electro-optic response was found to be highly sensitive to the fixed boundary orientations. We have not taken into account the effects of flow or of thermal fluctuations on the dynamics, effects which will be considered in future work.

We gratefully acknowledge the help of P. Willis and Z. Zhiming in this work, supported by NSF Engineering Research Center Grant No. CDR-862236 and NSF Solid State Chemistry Grant DMR 8901657.

[1] *Solitons in Liquid Crystals*, edited by L. Lam and J. Prost (Springer, New York, 1991).
[2] For a review see: L. Lei, S. Changqing, and X. Gang, J. Statis. Phys. **39**, 633 (1985).
[3] P. E. Cladis, H. R. Brand, and P. L. Finn, Phys. Rev. A **28**, 512 (1983).
[4] J. E. Maclenan, M. A. Handschy, and N. A. Clark, Phys. Rev. A **34**, 3554 (1986).
[5] J. E. Maclennan, N. A. Clark, and M. A. Handschy, in *Solitons in Liquid Crystals*, edited by L. Lam and J. Prost (Springer, New York, 1991), pp. 151–190.
[6] M. A. Handschy, N. A. Clark, and S. T. Lagerwall, Phys. Rev. Lett. **51**, 471 (1983).
[7] D. W. Berreman and T. J. Scheffer, Phys. Rev. Lett. **25**, 577 (1970).
[8] I. Abdulhalim, R. Weil, and L. Benguigui, J. Phys. **46**, 815 (1985).
[9] R. M. Sincovec and N. K. Madsen, A. C. M. Trans. Math. Software **1**, 232 (1975).