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The Electric Field in Polymer Dispersed Liquid Crystal Droplets and its Influence on Molecular Orientation

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We derived the expression for the electric field in an isolated spherical droplet of a uniformly oriented uniaxial liquid crystal contained within an isotropic medium, as a function of the dielectric and conductive properties of the materials and of the orientation of the liquid crystal. Results show that the field within the droplet is uniform and that its direction is different from the one of the applied field. Above a certain threshold field intensity the liquid director is reoriented but this simultaneously lowers the field in the droplet. This mechanism contributes to the lack of sharpness in the electrooptic response of PDLC's to an applied field and it must be considered together with the other already known mechanisms. This effect plays a negative role in multiplex addressing applications of PDLC films.

Keywords: Liquid crystals; PDLC

1. INTRODUCTION

Polymer Dispersed Liquid Crystals (PDLC) are a class of composite materials where liquid crystal droplets of controllable size and shape are dispersed within a solid matrix [1, 2]. Various kinds of liquid crystals can be used, as well as matrices of different nature, depending on the desired characteristics of the final material [3–5]. PDLC's are extremely interesting as probes for testing existing theories on liquid crystal/surface inter-

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actions [6−8], but they are even more interesting for their applications in optoelectronic devices [9]. In fact, in PDLC films, the anisotropic properties of liquid crystals allow for a reorientation of the mesophase director upon the application of an external field, and for the consequent variation of the film transparency.

Unlike in the traditional liquid crystal devices, the electric field in a nematic liquid crystal droplet surrounded by an isotropic medium, as in PDLC's, depends not only on the thickness of the PDLC cell and on the applied voltage, but also on the dielectric and conductive properties of the two materials. It would be convenient to express the distribution of the electric field by simple analytical functions and then use the continuum elastic theory to perform theoretical calculations of the director orientation and electrooptical behaviour of PDLC's. Such an approach presents several problems and until now only numerical calculations of the field have been performed [10]. On the contrary many experimental and theoretical studies exist on the director distribution within nematic or cholesteric droplets under the influence of an external field [11−15]. Several approaches have been used for the calculation of the minimum energy configuration but in all cases the field has been considered as independent of the configuration and homogeneous over the droplet volume.

Numerical results for the field, given a certain configuration, are suitable for further numerical combined calculations of the field and the configuration, but analytical expressions for the field would yield a better understanding of the role played by different factors. We performed an analytical calculation of the electric field present in an anisotropic spherical domain using a simplified but physically meaningful model based on the assumption that a uniform ac field is applied on a perfectly spherical isolated droplet surrounded by isotropic material. The liquid crystal in the droplet is assumed to be uniaxial and uniformly oriented in a monodomain, neglecting the elastic deformation near the interface. For such a system the expression of the field in the droplet, for any direction of the nematic symmetry axis, can be derived. This will allow us to know whether the field will be enhanced or reduced during the reorientation of liquid crystal molecules and prove that the variation of the field caused by the reorientation of the molecules contributes to the broadening of the electrooptical response to an external field. We believe that despite of simplification of the problem, the tendency predicted here represents one of the fundamental features of PDLC's.

The results of our calculations are relevant for understanding the nature of the optical response of PDLC's to an applied field. In fact, such films

typically are unresponsive to the field until its intensity reaches a certain threshold value. Above such value the optical properties of the film change gradually with increasing field intensity until a saturation value is reached, when optical properties are stable towards further field increase. It is interesting to investigate, also from the point of view of applications, the reasons for the graduality of the film optical response in the intermediate region. Until now two factors that contribute to lower the slope of the electrooptical response of PDLC's have been identified. One of them is connected with the distribution of droplets dimensions, since a different threshold field would be required to reorient average molecular directions in droplets with different size [11]. A second factor derives from the nature of the reorientation: it is clear that the field induced molecular reorientation in PDLC's is a process that is accomplished through a continuous evolution of the director distribution within the droplet, a process in which the effect of the field against the surface interactions is modulated by the elastic energy [13]. Since every director distribution corresponds to a different optical transmission, one gets a continuous change in the PDLC's transparency. One of the aims of this paper is to show that a third mechanism needs to be considered and that even if the threshold field could be the same for all droplets, still the slope of the electrooptical response would be less steep than expected, because for different orientations of the liquid crystal director, the internal field in the droplet is different.

2. THE MODEL

It would be rather complex to directly obtain an expression for the electric field in PDLC's from the conductivity and dielectric tensorial components, given a certain applied field. It is much simpler to calculate the distribution of electrical potential in the droplet and from this to obtain the expression for the field. Another difficulty is that, under the effect of the conductive tensor, charge will accumulate at the interface between the liquid crystal and the isotropic material. This effect will contribute to the time dependence of the field but it can be neglected if we restrict our attention to ac fields of high enough frequency, and in particular above the Maxwell-Wagner frequency [16]. Furthermore, we only treat the case when the frequency of the field is low enough to ensure that the electromagnetic wavelength be much larger than the dimension of the droplet. Such a consideration is realistic for liquid crystal display devices.

In the droplet, the field can be described by Maxwell's equation:

$$\nabla \cdot \left[\sigma \vec{E} + \frac{\partial}{\partial t} (\varepsilon \vec{E}) \right] = 0 \tag{1}$$

with σ and ε being the conductive and dielectric tensors respectively.

Let $\vec{E} = \vec{\mathbb{E}} e^{j\omega t}$, then:

$$\nabla \cdot [\sigma \vec{\mathbb{E}} + j\omega \varepsilon \vec{\mathbb{E}}] = 0 \tag{2}$$

Defining $\bar{\sigma} = \sigma + j\omega \varepsilon$ as the complex conductive tensor, Maxwell's equation can be written as:

$$\nabla \cdot \bar{\sigma} \vec{\mathbb{E}} = 0 \tag{3}$$

Referring to the principal cartesian axes (x, y and z) of the liquid crystal conductivity and dielectric tensors, assumed to be the same, with the z axis along the symmetry axis of the droplet, the tensor $\bar{\sigma}$ will have the diagonal form:

$$\bar{\sigma} = \begin{bmatrix} \sigma_{11} + j\omega\varepsilon_{11} & 0 & 0 \\ 0 & \sigma_{11} + j\omega\varepsilon_{11} & 0 \\ 0 & 0 & \sigma_{33} + j\omega\varepsilon_{33} \end{bmatrix} = \begin{bmatrix} \bar{\sigma}_{11} & & \\ & \bar{\sigma}_{11} & \\ & & \bar{\sigma}_{33} \end{bmatrix} \tag{4}$$

Defining $\vec{E} = -\nabla U$, where $U = V e^{j\omega t}$, as the potential, we get:

$$\bar{\sigma}_{11} \frac{\partial^2 V}{\partial x^2} + \bar{\sigma}_{11} \frac{\partial^2 V}{\partial y^2} + \bar{\sigma}_{33} \frac{\partial^2 V}{\partial z^2} = 0. \tag{5}$$

By applying the coordinate transformation to a new reference frame (x', y', z') with $x = x'$, $y = y'$ and $z' = \sqrt{\frac{\bar{\sigma}_{11}}{\bar{\sigma}_{33}}} z$, then

$$\frac{\partial^2 V}{\partial z^2} = \frac{\bar{\sigma}_{11}}{\bar{\sigma}_{33}} \frac{\partial^2 V}{\partial z'^2} \tag{6}$$

and Eq. (5) becomes, in the new (x', y', z') frame:

$$\bar{\sigma}_{11} \frac{\partial^2 V}{\partial x'^2} + \bar{\sigma}_{11} \frac{\partial^2 V}{\partial y'^2} + \bar{\sigma}_{11} \frac{\partial^2 V}{\partial z'^2} = 0. \tag{7}$$

This last expression is Laplace's equation: $\nabla'^2 V_- = 0$, where the subscript '−' indicates that this is the potential inside the droplet, while the

superscript' labels the new frame. Outside of the droplet the material is isotropic and Laplace's equation has the usual form in the old frame: $\nabla^2 V_+ = 0$, where the subscript $+$ indicates that the equation applies outside the droplet.

It is convenient to use spherical coordinates to proceed further. The new and old spherical coordinates are connected as follows:

$$\frac{r'^2}{r^2} = 1 - \left(1 - \frac{\bar{\sigma}_{11}}{\bar{\sigma}_{33}}\right)\cos^2\theta = 1 - \delta\cos^2\theta; \qquad (8a)$$

$$\cos\theta' = \sqrt{\frac{\bar{\sigma}_{11}}{\bar{\sigma}_{33}}}\frac{\cos\theta}{\sqrt{1 - \delta\cos^2\theta}}; \qquad (8b)$$

$$\sin\theta' = \frac{\sin\theta}{\sqrt{1 - \delta\cos^2\theta}}; \qquad (8c)$$

$$\phi' = \phi; \qquad (8d)$$

where $\delta = 1 - \frac{\bar{\sigma}_{11}}{\bar{\sigma}_{33}}$ is the anisotropy parameter.

At the interface of the droplet, two continuity conditions for the potential inside and outside the droplet must hold: the charge balance condition (see appendix)

$$\bar{\sigma}_0 \left.\frac{\partial V_+}{\partial r}\right|_{r=R} = \left(\frac{\partial V_-}{\partial r}(\bar{\sigma}_{11}\sin^2\theta + \bar{\sigma}_{33}\cos^2\theta)\right. \\ \left.+ \frac{\partial V_-}{\partial\theta}(\bar{\sigma}_{11} - \bar{\sigma}_{33})\frac{\sin\theta\cos\theta}{r}\right)\Bigg|_{r=R} \qquad (9)$$

and the potential continuity condition:

$$V_+|_{r=R} = V_-|_{r=R} \qquad (10)$$

3. ELECTRIC FIELD CALCULATION

As already mentioned, we consider here one isolated liquid crystal droplet surrounded by an isotropic material in an uniform applied electric field. In the following we describe the derivation of the distribution of the potential both in the droplet and in the isotropic material. In our principal coordinate system the symmetry axis of the director distribution within the droplet is set along the z axis, as shown in Figure 1. We choose our reference frame in
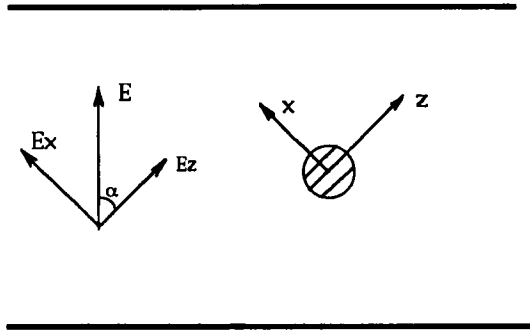
FIGURE 1   Reference frame used in field calculations.

such a way that the external field very far from the droplet (actually infinitely far from it) is defined in the *xz* plane and can then be split into two components, one being parallel to the *z* axis, another being parallel to the *x* axis. In the following the contributions to the potential due to such two external field components will be calculated and the total potential will then be obtained, from which the field can be easily derived.

A. The Potential Due to the *z* Component of the External Field

In this case the external uniform field is along the symmetry axis of the droplet, defined as the *z* axis. The field in the isotropic material will not be uniform near the interface because of the influence of the droplet, but far enough from the droplet the influence of the interface can be neglected and the field can be considered uniform. The potential at an infinite distance from the interface, $U_{+\infty z}$, can be expressed in the spherical reference frame as:

$$U_{+\infty z} = r E_0 \cos\theta \cos\alpha \, e^{j\omega t} = r E_0 P_1 (\cos\theta) \cos\alpha \, e^{j\omega t} \qquad (11)$$

or

$$V_{+\infty z} = r E_0 \cos\theta \cos\alpha = r E_0 P_1 (\cos\theta) \cos\alpha \qquad (12)$$

here $P_1(x)$ is the first order Legendre's polynomial, E_0 is the intensity of the external field and α is the angle between the *z* axis and the field. Equation (12) is an infinite boundary condition.

Outside of the droplet, Laplace's equation has a general solution in spherical coordinates [17]. When z is the symmetry axis, the potential in the surrounding isotropic material V_{+z} can be written as:

$$V_{+z} = \sum_{k=0}^{\infty} \left(A_k \, r^k + B_k \, \frac{1}{r^{k+1}} \right) P_k \left(\cos \theta \right) \tag{13}$$

Within the droplet, the potential V_{-z} is expressed, in the new frame, as:

$$V_{-z} = \sum_{k=0}^{\infty} \left(C_k \, r'^k + D_k \, \frac{1}{r'^{k+1}} \right) P_k \left(\cos \theta' \right) \tag{14}$$

here, A_k, B_k, C_k and D_k are expansion coefficients and $P_k(x)$ is the Legendre's polynomial of order k. Equations (13) and (14) are the general solutions of Laplace's equation for spherical polar coordinates in the case of a system with cylindrical symmetry. Using Eqs. (8), (9), (10) and (12), the coefficients can be found. The result is:

$$V_{z+} = -E_0 \, r \cos \alpha \cos \theta + \frac{\bar{\sigma}_{33} - \bar{\sigma}_0}{\bar{\sigma}_{33} + 2\bar{\sigma}_0} \, \frac{R^3}{r^2} \, E_0 \cos \alpha \cos \theta \tag{15}$$

$$V_{z-} = -\frac{3\bar{\sigma}_0}{\bar{\sigma}_{33} + 2\bar{\sigma}_0} \, E_0 \, r \cos \alpha \cos \theta \tag{16}$$

Subscripts $-$ and $+$ are used again to indicate the potential inside and outside the droplet, respectively, while $\bar{\sigma}_0$ is the conductivity outside the droplet. This result indicates that the potential is independent of $\bar{\sigma}_{11}$ when the external field is perpendicular to the xy plane.

B. The Potential Due to the x Component of the External Field

In the same frame used before, the potential at an infinite distance from the droplet associated with the x component of the field, $U_{+\infty x}$, which constitutes another infinite boundary condition, as:

$$U_{+\infty x} = r E_0 \sin \alpha \sin \theta \cos \phi \, e^{j\omega t} = r E_0 \sin \alpha \cos \phi \cdot P_1^1 \left(\cos \theta \right) e^{j\omega t} \tag{17}$$

or

$$V_{+\infty x} = r E_0 \sin \alpha \sin \theta \cos \phi = r E_0 \sin \alpha \cos \phi \cdot P_1^1 \left(\cos \theta \right) \tag{18}$$

here $P_1^1(x)$ is the first order associated Legendre polynomial, and other symbols have been defined before.

In this case z is not a symmetry axis and the general solution of Laplace's equation will be more complex. The potential outside the droplet, V_{+x}, will have the following form:

$$V_{+x} = \sum_{k=0}^{\infty} \left(\left(A_k r^k + B_k \frac{1}{r^{k+1}} \right) \sum_{m=0}^{k} (C_m \cos m\phi + D_m \sin m\phi) P_k^m (\cos\theta) \right),$$

(19)

while the potential inside the droplet V_{-x}, in the new frame, is:

$$V_{-x} = \sum_{k=0}^{\infty} \left(\left(F_k r'^k + G_k \frac{1}{r'^{k+1}} \right) \sum_{m=0}^{k} (H_m \cos m\phi' + I_m \sin m\phi') P_k^m (\cos\theta') \right).$$

(20)

Here A_k, B_k, C_m, D_m, F_k, G_k, H_m and I_m are expansion coefficients and $P_k^m(x)$ is an associated Legendre polynomial. As with Eqs. (13) and (14), Eqs. (19) and (20) are general solutions of Laplace's equation for spherical polar coordinates, but this time no restriction on the symmetry of the system is considered. Equations (8), (9), (10) and (18) can be used to evaluate the constants in (19) and (20). These solutions can be related to the ones for the z component of the external field, (15) and (16), and we may suppose that in the present case the potential contributions should be independent of $\bar{\sigma}_{33}$, and should have the same form as (15) and (16) respectively. Considering the expression for the infinite boundary condition (18), we try the following solution:

$$V_{+x} = - E_0 r \sin\alpha \sin\theta \cos\phi + \frac{\bar{\sigma}_{11} - \bar{\sigma}_0}{\bar{\sigma}_{11} + 2\bar{\sigma}_0} \frac{R^3}{r^2} E_0 \sin\alpha \sin\theta \cos\phi \quad (21)$$

$$V_{-x} = - \frac{3\bar{\sigma}_0}{\bar{\sigma}_{11} + 2\bar{\sigma}_0} E_0 r \sin\alpha \sin\theta \cos\phi \quad (22)$$

In fact, (21) and (22) can be written as:

$$V_{+x} = - E_0 r \sin\alpha \, P_1^1 (\cos\theta) \cos\phi + \frac{\bar{\sigma}_{11} - \bar{\sigma}_0}{\bar{\sigma}_{11} + 2\bar{\sigma}_0} \frac{R^3}{r^2} E_0 \sin\alpha \, P_1^1 (\cos\theta) \cos\phi$$

(23)

$$V_{-x} = - \frac{3\bar{\sigma}_0}{\bar{\sigma}_{11} + 2\bar{\sigma}_0} E_0 r' \sin \alpha \, P_1^1 (\cos \theta') \cos \phi \tag{24}$$

which are immediately recognized as solutions of Laplace's equation inside and outside the droplet, respectively. Substituting them into (9), (10) and (18) we find that they really are the solutions of our problem.

C. The Electric Field

The overall potential in the droplet is the sum of the potentials associated with both the z and the x external field components:

$$V_- = V_{-x} + V_{-z} = - \frac{3\bar{\sigma}_0}{\bar{\sigma}_{11} + 2\bar{\sigma}_0} E_0 x \sin \alpha - \frac{3\bar{\sigma}_0}{\bar{\sigma}_{33} + 2\bar{\sigma}_0} E_0 z \cos \alpha. \tag{25}$$

Thus we can get the expression for the total field:

$$\begin{aligned}
\vec{\mathbb{E}}_- = -\nabla V_- &= \frac{3\vec{\sigma}_0}{\bar{\sigma}_{11} + 2\bar{\sigma}_0} E_0 \sin \alpha \, \vec{x} + \frac{3\bar{\sigma}_0}{\bar{\sigma}_{33} + 2\bar{\sigma}_0} E_0 \cos \alpha \, \vec{z} \\
&= E_0 \left(\frac{3\bar{\sigma}_0}{\bar{\sigma}_{11} + 2\bar{\sigma}_0} \sin \alpha \, \vec{x} + \frac{3\bar{\sigma}_0}{\bar{\sigma}_{33} + 2\bar{\sigma}_0} \cos \alpha \, \vec{z} \right) e^{j\omega t}
\end{aligned} \tag{26}$$

The corresponding expression for the field in the surrounding material could be obtained using the same method, but since it is beyond our purposes we will omit its derivation here.

4. RESULTS AND CONCLUSIONS

Figure 1 shows the relative orientation of the external field with respect to the symmetry axis of the droplet director distribution. From Eq. (26) we can immediately reach the following conclusions:

1. The $\vec{E}_-$ vector is independent of position and droplet radius. This indicates that within all PDLC droplets the field is uniform and its intensity is the same regardless of the droplet radius.
2. The angle β between the field in the droplet and the z axis is given by:

$$\tan \beta = \frac{|\bar{\sigma}_{33} + 2\bar{\sigma}_0|}{|\bar{\sigma}_{11} + 2\bar{\sigma}_0|} \tan \alpha \tag{27}$$

The field in the droplet and the external field would be parallel only when $\alpha = 0$, $\pi/2$ or $\bar{\sigma}_{33} = \bar{\sigma}_{11}$, that is to say, when the direction of the applied field is parallel to one of the principle axes, or when the droplet becomes isotropic.

3. Since $\bar{\sigma}_i = \sigma_i + j\omega\varepsilon_i$ $(i = 0, 11, 33)$ are complex numbers, unless $\sigma \gg \varepsilon_i$ or $\sigma_i \ll \varepsilon_i$, the phase of the field in and out of the droplet is different.

4. In applications of PDLC's, the condition $\sigma_i \ll \varepsilon_i$ is desired, both to extend its working life and to reduce power consumption. In this case the intensity of the field can be written in the following form:

$$|\vec{E}| = E_0 \left(\left(\frac{3\varepsilon_0}{\varepsilon_{11} + 2\varepsilon_0} \sin\alpha \right)^2 + \left(\frac{3\varepsilon_0}{\varepsilon_{33} + 2\varepsilon_0} \cos\alpha \right)^2 \right)^{1/2} \qquad (28)$$

A typical electroptic response of a PDLC film to an external field is shown in Figure 2. Below a certain value of the field, which we call the threshold field, there is no change in the material optical transparency. Above such threshold the transparency increases gradually with the field until it reaches a plateau. The transparency variation is of course associated with molecular reorientation within the droplet. When the field in the droplet is just above the threshold field and $\varepsilon_{33} > \varepsilon_{11}$, molecuels will begin to turn under the effect of the field, and α becomes smaller. Within the frame of our model we see from Eq. (26) that with the decreasing of α, the field in the droplet would decrease. The reorientation of the molecules, which in our simple model
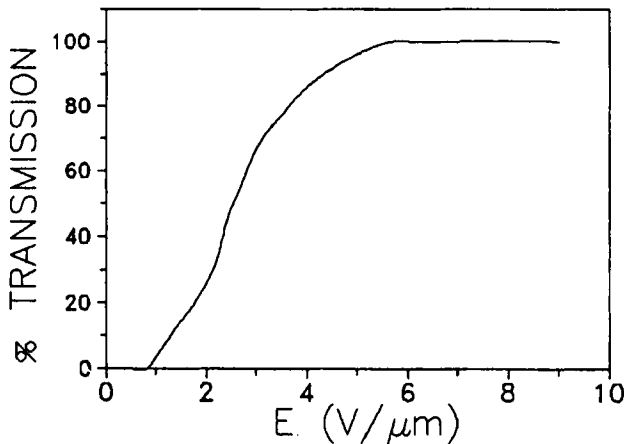

FIGURE 2   A typical experimental optical response of a PDLC film to an electric field.

starts at the threshold field, would then stop immediately when the threshold field is reached. In other words, the director will turn until the field in the droplet decreases to the value of the threshold field E_{th}. The angle between the director and the external field α can be obtained from:

$$E_0\left(\left(\frac{3\varepsilon_0}{\varepsilon_{11}+2\varepsilon_0}\sin\alpha\right)^2 + \left(\frac{3\varepsilon_0}{\varepsilon_{33}+2\varepsilon_0}\cos\alpha\right)^2\right)^{1/2} = E_{th} \qquad (29)$$

for $\varepsilon_{33} < \varepsilon_{11}$, molecules will tend to orient perpendicularly to the field, but also in this case the reorientation of the director is associated with a decreasing intensity of the field in the droplet and the reorientation of the liquid crystal director stops at the threshold field.

Figures 3 and 4 show the dependence of the angle α, which defines the orientation of the liquid crystal director with respect to the applied field (see Fig. 1), on the intensity of the applied field E_0 for different values of the dielectric properties of the materials. In all cases such values have been chosen within the range of what is usually experimentally measured. We can see in particular, from Figure 3, how the sharpness of the reorientation dramatically increases when the matrix dielectric constant increases. Figure 4, on the other hand, shows how the sharpness increases with decreasing dielectric anisotropy of the liquid crystal. Such information can be extremely valuable in designing PDLC performance when these materials are used for specific applications.

In our model only one of the causes that contribute to the lack of sharpness of the reorientation process has been taken into consideration,
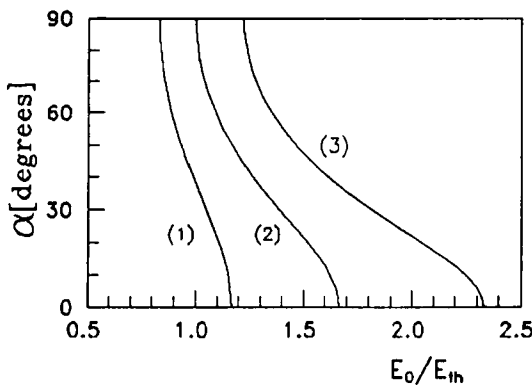

FIGURE 3   The dependence of liquid crystal orientation on the applied field, as obtained from Eq. (29), for (1) $\varepsilon_0 = 10$, (2) $\varepsilon_0 = 5$ and (3) $\varepsilon_0 = 3$. In all cases $\varepsilon_{11} = 5$ and $\varepsilon_{33} = 15$.
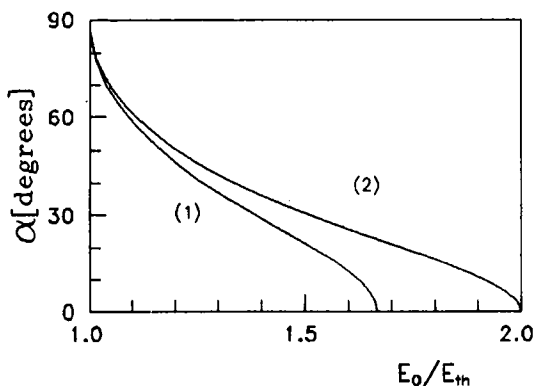
FIGURE 4 The dependence of liquid crystal orientation on the applied field, as obtained from Eq. (29), for different values of liquid crystal anisotropies. In particular $\varepsilon_{33} = 15$ for curve (1) and $\varepsilon_{33} = 20$ for curve (2). In both cases $\varepsilon_0 = \varepsilon_{11} = 5$.

namely the variation of the field intensity within the droplet which is consequent to the reorientation itself. This model is crude when it tries to represent the reorientation process, since it neglects several factors like the droplet size dispersion or the ones connected to the elastic deformations. Nevertheless it has the advantage of making us understand the contribution of a single mechanism to the sharpness of the reorientation.

The variation of the field seen by the liquid crystal with its orientation is a feature typical of PDLC's and it does not exist in most other types of liquid crystal materials. Only as a consequence of the dielectric properties of the materials, the multiplexing properties of displays based on PDLC's can then be quite discouraging, and things can get only worse if we consider other factors which contribute to the lack of sharpness of the electrooptic response, like the droplet size distribution or the elastic nature of the reorientation. Of course non-commercial liquid crystals and polymers which may have more suitable dielectric character could be chosen.

In our derivation, only one droplet was taken into consideration but the presence of neighbouring droplets might influence the internal field in such a way that the severe limitations of dielectric parameters would be reduced. Work is in progress to explore such possibilities.

Acknowledgements

Fine II. Discussions with professor L. Komitov were extremely useful to the authors.

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APPENDIX

At the interface of the droplet, their charge balance condition can be expressed, for fields above the Maxwell-Wagner frequency, as:

$$\nabla \cdot [\bar{\sigma}\vec{\mathbb{E}}] = 0 \qquad (A1)$$

that is,

$$[\bar{\sigma}\vec{\mathbb{E}}_-]_n|_{r=R} = [\bar{\sigma}_0\vec{\mathbb{E}}_+]_n|_{r=R}. \qquad (A2)$$

here n indicates the direction normal to the interface, R is the radius of the droplet and $\bar{\sigma}$ and $\bar{\sigma}_0$ are the complex conductivities inside and outside the droplet, respectively. At the surface of the spherical droplet the normal direction is just the direction of the r axis. The left side of the above equation must be considered in the droplet, where we have an anisotropic material.

We can then write:

$$\vec{\mathbb{D}}_- = [\bar{\sigma}\vec{\mathbb{E}}]_- = \begin{bmatrix} \bar{\sigma}_{11}E_x \\ \bar{\sigma}_{11}E_y \\ \bar{\sigma}_{33}E_z \end{bmatrix} = \begin{bmatrix} D_{x-} \\ D_{y-} \\ D_{z-} \end{bmatrix} \tag{A3}$$

in Cartesians coordinates, or:

$$\mathbb{D} = \begin{bmatrix} D_r \\ D_\theta \\ D_\phi \end{bmatrix} = \begin{bmatrix} \frac{\partial r}{\partial x} & \frac{\partial r}{\partial y} & \frac{\partial r}{\partial z} \\ \frac{\partial \theta}{\partial x} & \frac{\partial \theta}{\partial y} & \frac{\partial \theta}{\partial z} \\ \frac{\partial \phi}{\partial x} & \frac{\partial \phi}{\partial y} & \frac{\partial \phi}{\partial z} \end{bmatrix} \cdot \begin{bmatrix} D_{x-} \\ D_{y-} \\ D_{z-} \end{bmatrix} \tag{A4}$$

in spherical coordinates. We then have:

$$D_{r-} = \bar{\sigma}_{11}E_{x-}\frac{\partial r}{\partial x} + \bar{\sigma}_{11}E_{y-}\frac{\partial r}{\partial y} + \bar{\sigma}_{33}E_{z-}\frac{\partial r}{\partial z} \tag{A5}$$

with:

$$-E_{x-} = \frac{\partial V_-}{\partial x} = \frac{\partial V_-}{\partial r}\frac{\partial r}{\partial x} + \frac{\partial V_-}{\partial \theta}\frac{\partial \theta}{\partial x} + \frac{\partial V_-}{\partial \phi}\frac{\partial \phi}{\partial x} \tag{A6}$$

$$-E_{y-} = \frac{\partial V_-}{\partial y} = \frac{\partial V_-}{\partial r}\frac{\partial r}{\partial y} + \frac{\partial V_-}{\partial \theta}\frac{\partial \theta}{\partial y} + \frac{\partial V_-}{\partial \phi}\frac{\partial \phi}{\partial y} \tag{A7}$$

$$-E_{z-} = \frac{\partial V_-}{\partial z} = \frac{\partial V_-}{\partial r}\frac{\partial r}{\partial z} + \frac{\partial V_-}{\partial \theta}\frac{\partial \theta}{\partial z} + \frac{\partial V_-}{\partial \phi}\frac{\partial \phi}{\partial z} \tag{A8}$$

here the subscript "_" indicates that the calculation is performed in the droplet. We then have:

$$[\bar{\sigma}\vec{\mathbb{E}}]_{n-} = D_{r-} = -\frac{\partial V_-}{\partial r}(\bar{\sigma}_{11}\sin^2\theta + \bar{\sigma}_{33}\cos^2\theta) - \frac{\partial V_-}{\partial \theta}(\bar{\sigma}_{11} - \bar{\sigma}_{33})\frac{\sin\theta\cos\theta}{r} \tag{A9}$$

inside the droplet, while outside the droplet:

$$[\bar{\sigma}\vec{\mathbb{E}}]_{n+} = -\bar{\sigma}_0\frac{\partial V_+}{\partial r}. \tag{A10}$$

The charge balance condition is then:

$$\bar{\sigma}_0\frac{\partial V_+}{\partial r}\bigg|_{r=R} = \left(\frac{\partial V_-}{\partial r}(\bar{\sigma}_{11}\sin^2\theta + \bar{\sigma}_{33}\cos^2\theta) + \frac{\partial V_-}{\partial \theta}(\bar{\sigma}_{11} - \bar{\sigma}_{33})\frac{\sin\theta\cos\theta}{r}\right)\bigg|_{r=R} \tag{A11}$$