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PDLC shutters: where has this technology gone?

J. William Doane[a]; Bao-Gang Wu[b]; John H. Erdmann[b]; J. William Doane[b]
[a] Kent Displays, Inc., 343 Portage Blvd., Kent, OH 44240 [b] Liquid Crystal Institute and Department of Physics, Kent State University, Kent, Ohio 44242, U.S.A

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PDLC shutters: where has this technology gone?

J. WILLIAM DOANE*
Kent Displays, Inc., 343 Portage Blvd., Kent, OH 44240, USA

A Commentary on the paper "Response times and voltages for PDLC shutters", by Bao-Gang Wu, John H. Erdman and J. William Doane. First published in *Liquid Crystals*, **5**, 1453-1465 (1989).

The history behind this work started from an accident that occurred in the laboratory when one of the authors, J.W. Doane of Kent State University, was working with Professor G. Chidichimo of Calabria University attempting to seal a nematic liquid crystal material in a vial with a two-part epoxy glue. Before the clear epoxy mixture had time to cure, it accidentally flowed into the liquid crystal making a big mess on the laboratory bench. Being late in the day, the two investigators decided to go home and leave the mess until the next day. When they returned in the morning, much to their surprise, the epoxy/liquid crystal mixture had turned from a clear liquid into a beautiful white rubbery solid. Upon further study of the white solid they discovered that, in the curing process, the liquid crystal had separated from the epoxy to form micron size droplets nicely sealed and uniformly distributed inside the cured epoxy. The nematic liquid crystal inside the droplets was surprisingly pure offering a new way to study nematics confined to a small cavity. Furthermore, it was found that the material could be electrically switched from a translucent white to an optically clear appearance offering a new form of a light shutter or electronic display device. The effect of an applied electrical field was to align the nematic director of the droplets in a direction such that their refractive index effectively matched that of the epoxy polymer thereby removing the light scattering property and changing the material from a white opalescence to optically clear. Upon searching the literature we found that such an index switching phenomenon had actually been predicted and patented in England, by C. Hilsum [1] although he had not envisioned making such a device this way.

We subsequently referred to this material as a polymer dispersed liquid crystal [2], PDLC, a term that was later extended to include other types of liquid crystal dispersions made from waterborne emulsions, an entirely different process [3]. This method of emulsion processing was introduced much earlier in the 1960s and first used as a method to encapsulate and disperse chiral liquid crystals for temperature sensing; still manufactured, even to this day, as thermometers primarily used in medically related applications.

This particular paper in *Liquid Crystals* by my students John Erdmann and B-G.Wu and I arose from our attempt to understand better the electrooptic response of a PDLC. At that time there was an intense effort by us and many others to develop them for applications in such devices as electronic displays and switchable windows [2, 3]. For these applications, it is important to be able to design PDLC materials to optimize or select drive voltages as well as response times, depending upon the application. This paper was a simple approach to understanding the effects of droplet morphology as well as the dielectric permittivity and conductivity on the material response.

As it turns out, PDLC materials were indeed commercialized by several companies for window applications; however, because of the high cost of nematic liquid-crystalline material and of the window substrates containing transparent electrodes, the window market has been kept small. Nevertheless magnificent switchable windows were and are still made for offices and homes. The manufactured windows have simply been too expensive, however, to be used in high volume markets such as windows in the average family home. PDLC materials were also developed for light scattering displays using the index match/mismatch method. This application has been even less successful than windows in the market place. The principal reason for this is because the light scattering white texture in not nearly scattering enough for the white texture to be as bright as paper. Similarly, the clear texture in not clear enough so that there is not sufficient contrast to show an image as good as ink on paper. There have, however, been clever means developed to improve the contrast such as the incorporation of dyes [3] but a

*Email: jwdoane@lci.kent.edu or bdoane@kentdisplays.com

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high volume PDLC display product using the index matching method has yet to be found.

It is, perhaps, because of the considerable activity in the display and window technology that this paper was widely read. It should be mentioned that there has been considerable effort world wide to use PDLC materials as a means to study fundamental effects of surfaces and confinement on nematics and other liquid crystals. The large surface-to-volume ratio creates new physical effects that are extremely interesting to study. One author, John Erdmann, dedicated much of his Ph.D. dissertation to understanding the different director configurations that can occur in a droplet and measuring surface and confinement parameters that affect them [4].

Quite interestingly, the major commercial display application of PDLCs at present is not with nematic liquid crystals but with the chiral version of nematics, namely chiral nematic liquid crystals [5]. These materials disperse as easily as regular nematics either by phase separation or by the emulsion process; however, their electrooptic features are quite different. These materials provide iridescent colours due to Bragg reflection from the chiral induced helical twist. Of more importance for display applications, the chiral nematic dispersions are bistable offering an extremely low power, reflective display; a unique electrooptic curve further allows the production of a high resolution display without an expensive active matrix. Because polymer dispersions can be coated and printed, displays are being developed and produced on flexible materials such as plastic [6], paper and even fabric [7]

The authors are grateful for the opportunity to comment on this paper long after it was written. We hope that it has in some way given guidance to the *Liquid Crystals* readership.

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Response times and voltages for PDLC light shutters

BAO-GANG WU†, JOHN H. ERDMANN and J. WILLIAM DOANE*
Liquid Crystal Institute and Department of Physics, Kent State University, Kent, Ohio 44242, U.S.A

The response times and operating voltages of light shutters formed from polymer dispersed liquid crystals (PDLCs) have been studied experimentally and the results compared with calculations based on non-sperhically shaped nematic droplet models. The experiments were performed on light shutters with elongated and uniformly aligned droplets where the relaxation time and voltage response were measured. It is shown that the droplet shape can be a dominant factor, particularly for the relaxation time, and the data are compared with equations derived in terms of the aspect ratio of the droplet $l=a/b$, where a and b are the lengths of the semi-major and semi-minor axes, respectively, of the elongated droplet. It is further demonstrated that the electric field inside the droplet can be considerably smaller than the applied field, due to the conductivity and dielectric properties of the polymer and liquid crystal materials. These data are used to obtain values for the ratio of the conductivities of the polymer binder and liquid crystal droplet, as well as the anisotropy of the conductivity in the liquid crystal.

1. Introduction

Polymer dispersed liquid crystals (PDLCs) are a new type of light shutter which have been explored in recent years [1–4] and are beginning to find applications in switchable windows, displays, colour projectors and other devices [5]. These materials consist of submicrometre size nematic droplets dispersed in a polymer matrix, and their optical response is based on the electrically controlled light scattering properties of the droplets. An applied electric field aligns the nematic droplets to yield a transparent or non-scattering state. Surface interactions at the droplet wall and the droplet shape return the droplets to their original orientation in the absence of the field to yield a scattering or opaque state. A competiton between the applied field and the elastic and viscous torques of the liquid crystal govern the response times and switching voltages of the light shutters.

A typical light shutter prepared with droplets about 1.0 μm in diameter requires a switching field of approximately $1.0\,\mathrm{V\,\mu m^{-1}}$ and has a response of about 100 ms upon removal of the voltage. There can, however, be large departures from this. For example, a film prepared from epoxy polymers can sometimes have a response time of near 1.0 s. Stretching the film or applying a shear stress to the glass substrates elongates the droplets and shortens the response time to <1.0 ms without a substantial increase in the voltage threshold required to switch the film. Reducing the size of the droplets can have the same effect. The type of polymer binder can also affect the voltage response. For example, a light shutter prepared from poly(methyl methacrylate) (PMMA) can have a switching voltage of 200 V, whereas an identical shutter with identical droplet sizes and shapes prepared from an epoxy polymers can have a switching voltage of 20 V.

Here we report an experimental and theoretical study to explain and quantify some of these features in PDLC light shutters. In this study particular emphasis is placed upon the effects of droplet shape on the shutter response. In droplets with a particular type of nematic director configuration, namely the bipolar configuration [5], droplet shape can be a significant factor. Experiments have been designed in which the droplets are elongated and aligned by shearing the glass substrates of the shutter. The optical response characteristics were then measured using laser light, and were compared with model calculations. Models of two different droplet shapes with small and with large radii of curvature are compared with measurement of the response time with reasonable success, and the results are instructive. The voltage response is more difficult to model as it is found that the conductivity and dielectric properties of the polymer and liquid crystal play a strong role. Again, some preliminary models are presented with reasonable fits to experiment. These studies indicate that the ratio of conductivities of the polymer and liquid crystal is significant in the switching voltage.

*Corresponding author. Email: jwdoane@lci.kent.edu or bdoane@kentdisplays.com
† Present address: Polytronix, Inc., P.O. Box 833024, Richardson, Texas 75083, U.S.A.

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We believe this work to be only the beginning in understanding the electro-optic response of PDLC films. Significant future experimentation and theory are required to understand this subject.

2. Model calculations

Under static conditions the director configuration inside a droplet is, in part, understood under conditions where the droplet is large (>1.0 μm in diameter) and strong anchoring conditions apply. For spherical droplets, computer simulations have been found to predict reasonably well those configurations observed by optical microscopy [6–9]. Recently we have obtained computer simulations of director configurations which result under the conditions of an applied field and also for droplets which have the shape of an ellipsoid of revolution [10]. Figure 1 shows such a computer simulation of the bipolar configuration, which is one case that results from tangential wall alignment and the most common configuration that we have observed in PDLC materials. Because of the tangential anchoring at the polymer surface, application of an electric field aligns the bipolar axis but appears to have minimal effect on the bipolar configuration of the droplet, as illustrated by the computer simulation in figure 1 (*b*). For a perfectly spherical polymer cavity, little elastic distortion is required to align a bipolar droplet by an electric field to preserve the tangential anchoring at the cavity wall. In fact, the question arises of how a spherical bipolar droplet would reorient to its original orientation upon removal of an applied field. A possible answer is that in practice the droplets in PDLC materials are never perfectly spherical. The polymer binder is often a soft material and the droplet cavities easily become elongated or flattened during the fabrication of the shutter or during the phase separation process. Differences between the thermal expansivities of the polymer and glass or plastic cell substrate can also shape the droplets.

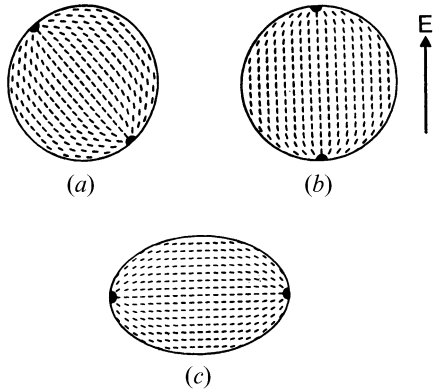

Figure 1. The bipolar configuration obtained from tangential anchoring (*a*) in the absence of a field, (*b*) in the presence of a strong electric field, and (*c*) in an elongated cavity. Three dimensional patterns can be obtained by rotation of these figures about the symmetry axis.

It is possible to model and calculate the effect of droplet shape on the response time of a PDLC light shutter with bipolar configured droplets. This is done by recognizing that the deformation free energy density inside the droplet is governed by the radius of curvature of the cavity. In a droplet which is slightly elongated in shape the bipolar axis will prefer to orient along the long axis of the cavity (see figure 1 (*c*)) where both splay and bend deformations are minimized. At the equator of the elongated bipolar droplet, near the wall, the elastic deformation is pure bend and the free energy density is $K_{33}/2R^2$, where K_{33} is the bend constant and R is the radius of curvature at the equator.

We generalize this concept to model the free energy density, F_d, of a nematic droplet in an elongated cavity, by using the expression $F_d = (K/2)\varrho^2$, where ϱ is the radius of curvature at the cavity wall indicated by the position vector, **r**, in figure 2 (*a*) and K is an effective deformation constant. Ignoring the details of the director configuration, we assign a droplet director, **n**, shown in figure 2 (*a*), which can reorient in the elongated cavity. In analogy to the bipolar droplet, the droplet director has its preferred orientation in the direction of the long axis of the cavity in the absence of a field. This simplified model assumes that we can approximate the dynamics of a droplet as one in which the droplet director reorients upon the onset or removal of a field with little variation in the effective value of K. The elastic restoring torque, Γ_d, per unit volume is given as

$$\Gamma_d = \mathrm{d}F_d/\mathrm{d}\lambda, \tag{1}$$

where the functional form of Γ_d depends upon the shape of the droplet and λ, the angle between the applied electric field, **E**, and the droplet director, **n**, as shown in figure 2 (*a*). Later we apply equation (1) to two specific droplet shapes, which exhibit a small and large radii of curvature.

The electrostatic free energy density inside the droplet due to an applied field is given by

$$F_e = -\frac{\varepsilon_\perp E^2}{2} - \frac{\Delta\varepsilon(\mathbf{n}\cdot\mathbf{E})^2}{2}, \tag{2}$$

where $\Delta\varepsilon = \varepsilon_\parallel - \varepsilon_\perp$ is the anisotropy in the dielectric constant. Application of the electric field serves to rotate **n** to its equilibrium orientation **N**. The orientation of **N** approaches the direction of **E** as the field

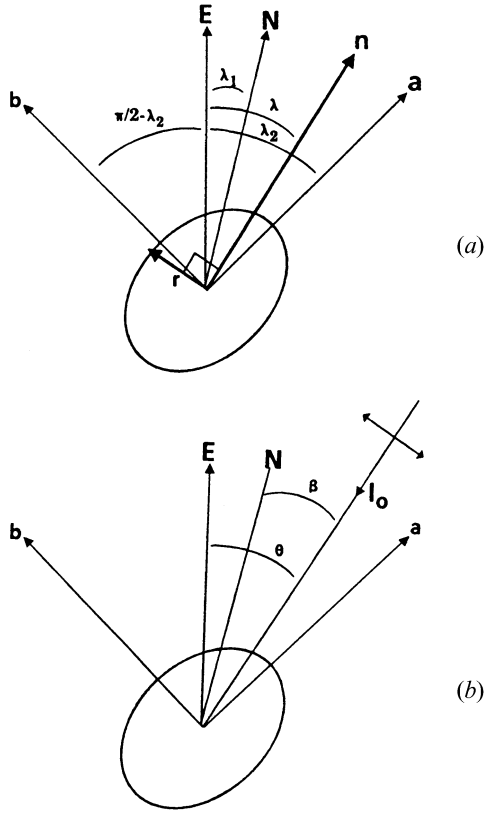

Figure 2. (*a*) Illustration of the droplet director, **n**, arbitrarily oriented in an elongated cavity where **r** is the position vector (perpendicular to the director) indicating the point on the wall where the radius of curvature is of interest. The vector **N** is the equilibrium orientation of the director in an electric field, **E**, of arbitrary strength. Directions **a** and **b** are the orientation of the major and minor axes of the elongated droplet. (*b*) Illustration of an incident polarized light of intensity, I_0 at an angle β from the ON state droplet director, **N**, and angle θ from the direction of the electric field **E**.

strength is increased, provided $\Delta\varepsilon$ is positive. Assuming a uniform field in the droplet, the electric torque on the droplet director, Γ_e, per unit volume is given by

$$\Gamma_e = \frac{dF_e}{d\lambda} = \frac{\Delta\varepsilon}{2}(E)^2 \sin 2\lambda. \tag{3}$$

A difficult problem is to relate the field inside the droplet to the applied field V/d_0, where V is the voltage applied to the transparent conducting electrodes of a light shutter with a PDLC film thickness of d_0. An exact calculation is beyond the scope of this paper, but simple and crude approximations are possible. To account for the effects of the boundary between the polymer and the nematic droplet, we assume an isolated spherical droplet in a polymer which is in a uniform electric field. Application of electrostatic boundary conditions in a one droplet approximation yields the field, E_2, inside the droplet as [11]

$$E_2 = E_1 \frac{3\varepsilon_1}{2\varepsilon_1 + \varepsilon_2} \underset{(\omega \to 0)}{=} 3E_1 \left(\frac{\sigma_2}{\sigma_1} + 2\right)^{-1}, \tag{4}$$

where E_1 is the field in the polymer and ε_1 and ε_2 the dielectric constants of the polymer and liquid crystal, respectively. We take ε_i to be complex where $\varepsilon_i = \varepsilon_i' + j\sigma_i/\omega$ and σ_i is the conductivity, ω is the angular frequency and ε_i' is the real part of the susceptibility. In the zero frequency (D.C.) limit, the ratio of the conductivities can govern the magnitude of the field inside the droplet. The value of σ_2 is known to be anisotropic [12]. We account for this anisotropy by

$$\sigma_2 = \sigma_\parallel \sigma_\perp \left(\sigma_\parallel \sin^2 \lambda, + \sigma_\perp \cos^2 \lambda\right)^{-1}, \tag{5}$$

where $\sigma_\parallel$ and $\sigma_\perp$ are the conductivities parallel and perpendicular, respectively, to the droplet director. Equation (5) assumes uniform director orientation within the droplet. In our experiments D.C. pulses were applied and so we are interested in the field within the droplet after the D.C. voltage has been applied for a sufficient length of time for the droplet director to achieve its equilibrium orientation, **N**, at an angle λ_1 as defined in figure 2 (*a*).

A viscous torque, Γ_v, opposes reorientation of the droplet director. In the absence of flow in a nematic material, this torque can be expressed by

$$\Gamma_v = \gamma_1 \frac{d\lambda}{dt}, \tag{6}$$

where γ_1 is the rotational viscosity coefficient.

We now consider elongated droplets of two different shapes.

2.1. *Model A (small radius of curvature)*

In this model we consider an elongated droplet where the radius of curvature at the droplet edge is

$$\varrho = a\left[\cos^2(\lambda_2 - \lambda) + l^2 \sin^2(\lambda_2 - \lambda)\right]^{-1/2}, \tag{7}$$

where a and b are the lengths of the semi-major and semi-minor axes, respectively, $l = a/b$ is the aspect ratio of the droplet, and λ_2 is the orientation of the long axis of the droplet and is defined in figure 2 (*a*). Applying equation (1), the elastic torque, Γ_d, per unit volume becomes

$$\Gamma_d = \frac{dF_d}{d\lambda} = -\frac{K(l^2 - 1)}{2a^2} \sin 2(\lambda_2 - \lambda). \tag{8}$$

2.1.1. Field response. We balance the elastic and field torques to determine the equilibrium orientation **N** is the presence of a voltage applied to a shaped droplet. Setting $\Gamma_e+\Gamma_d=0$,

$$\lambda_1=\frac{1}{2}\arctan\left(\frac{\sin 2\lambda_2}{A+\cos 2\lambda_2}\right), \tag{9}$$

where $A=\Delta\varepsilon a^2E^2/K(l^2-1)$. An interesting condition results at $\lambda_2=\pi/2$, where there is a critical voltage

$$V_c=\frac{d_0}{3a}\left(\frac{\sigma_2}{\sigma_1}+2\right)\left(\frac{K(l^2-1)}{\Delta\varepsilon}\right)^{1/2}. \tag{10}$$

This can be regarded as the shape contribution to the switching voltage of a normal PDLC shutter. This may, in fact, be the dominant contribution for a PDLC shutter as illustrated by applying equation (10) to a shutter prepared from epoxy materials wherein the normal process of fabricating the cell droplets may become slightly deformed, say $l=1.1$. A shutter made from epoxy materials commonly has a switching voltage of $V_c\approx 30\,\mathrm{V}$ for $d_0=25\,\mu\mathrm{m}$ and $a=1.0\,\mu\mathrm{rn}$, where cyanobiphenyl liquid crystals are used with $\Delta\varepsilon=14\varepsilon_0$ and $K\approx 1\times 10^{-11}\,\mathrm{N}$. Application of equation (10) yields $\sigma_2/\sigma_1\approx 25$, which is not unreasonable in view of the fact that Chidichimo *et al.* [13] have reported experimental evidence for dramatic reductions in the switching voltage as the conductivity of the polymer, σ_1, is increased. We have therefore ignored contributions other than the shape, such as changes in the director configuration or other factors toward estimating the voltage response of the shutter.

2.1.2. Relaxation time. When the electric field is removed, the orientation of the droplet director returns from an angle of λ_1 to λ_2. To calculate the relaxation time we balance the elastic and viscous torques ($\Gamma_d+\Gamma_v=0$), ignoring the inertial term, to obtain

$$\lambda=\lambda_2-\arctan\left\{\tan(\lambda_2-\lambda_1)\exp\left[-\left(\frac{K(l^2-1)t}{\gamma_1 a^2}\right)\right]\right\}. \tag{11}$$

The time constant for the relaxation time is therefore

$$\tau_{off}=\frac{\gamma_1 a^2}{K(l^2-1)}. \tag{12}$$

The effect of shaping on the relaxation time is seen to be appreciable, as estimated by taking $\gamma_1=4\times 10^{-2}\,\mathrm{kg\,m^{-1}\,s^{-1}}$ and the previous values for the other parameters. A slightly shaped droplet of $l=1.1$ yields $\tau_{off}\approx 20\,\mathrm{ms}$. This is comparable with that observed in weakly stretched films. The small value of l illustrates the large effect droplet shape can have on the relaxation time. It is noted that equation (12) accounts only for the droplet shape. In the case of a spherical droplet, $l=1$, the relaxation time is undefined since surface interactions and configuration changes were ignored.

2.1.3. Response time. The response time upon application of the electric field is calculated by balancing all of the torques; $\Gamma_e+\Gamma_d+\Gamma_v=0$ which yields

$$\lambda=\arctan\left[\frac{1}{\tan\lambda_1}\left(\frac{1+\tan^2\lambda_1}{1-m\exp(-t/\tau_{on})}-1\right)\right], \tag{13}$$

with λ_1 given by equation (9),

$$m=\frac{\tan\lambda_1\tan\lambda_2-\tan^2\lambda_1}{\tan\lambda_1\tan\lambda_2+1} \tag{14}$$

and

$$\tau_{on}^{-1}=\frac{1}{\gamma_1}\left(\Delta\varepsilon E^2+\frac{K(l^2-1)}{a^2}\frac{1-\tan^2\lambda_2}{1+\tan^2\lambda_2}\right)\left(\frac{1+\tan^2\lambda_1}{1-\tan^2\lambda_1}\right). \tag{15}$$

For $\lambda_2\approx\pi/2$ and $\lambda_1\approx 0$

$$\tau_{on}^{-1}=\left[\frac{1}{\gamma_1}\left(\Delta\varepsilon E^2+\frac{K(l^2-1)}{a^2}\right)\right] \tag{16}$$

and for large voltages $\tau_{on}=\gamma_1/\Delta\varepsilon E^2$.

2.2. *Model B (large radius of curvature)*

In order to investigate further the effect of droplet shape, we have selected another model, an ellipse, which provides a larger radius of curvature following

$$\varrho=\frac{a}{l^2}\left(\frac{l^4+\tan^2(\lambda_2-\lambda)}{l^2+\tan^2(\lambda_2-\lambda)}\right)^{3/2}. \tag{17}$$

The aspect ratio of the ellipse is made by a cross-sectional cut in the plane of the largest and smallest axes of the spheroid. The restoring torque, Γ_d, is

$$\Gamma_d=\frac{dF_d}{d\lambda}=-\frac{3Kl^6(l^2-1)}{a^2}\frac{\tan(\lambda_2-\lambda)\left[l^2+\tan^2(\lambda_2-\lambda)\right]^2}{\cos^2(\lambda_2-\lambda)\left[l^4+\tan^2(\lambda_2-\lambda)\right]^4}. \tag{18}$$

Note that for $l=1$, a spherical droplet, there is no restoring torque and $a=r_{droplet}$. Also, for $\lambda_2=\lambda$ the elastic deformation energy is minimized.

2.2.1. Voltage response. Upon application of an electric field, **n** moves away from the OFF state position. For small fields $\lambda_1=\lambda_2$ and for very large fields $\lambda_1\to 0$. After equilibrium is established in the ON state ($\lambda=\lambda_1$), there is a static balance between the electric and the elastic deformation torques with the

resulting expression

$$V=\frac{d_0 l^3}{a}\frac{\left[(\sigma_\parallel/\sigma_1)+2\right]}{\cos(\lambda_2-\lambda_1)}\frac{l^2+\tan^2(\lambda_2-\lambda_1)}{\left[l^4+\tan^2(\lambda_2-\lambda_1)\right]^2}$$
$$\left[\frac{3\varepsilon_0\sin 2\lambda_1}{2K(l^2-1)\tan(\lambda_2-\lambda_1)}\times\right. \tag{19}$$
$$\left.\left(\Delta\varepsilon-\frac{2(\varepsilon_\perp+\Delta\varepsilon\cos^2\lambda_1)(\sigma_\parallel/\sigma_1)\left[(\sigma_\parallel/\sigma_\perp)-1\right]}{\left[(\sigma_\parallel/\sigma_1)+2\right]\left[(\sigma_\parallel/\sigma_\perp)\sin^2\lambda_1+\cos^2\lambda_1\right]}\right)\right]^{-1/2},$$

where σ_1 is the conductivity of the polymer binder.

2.2.2. Relaxation time. In this case a balance of the viscous and electric torques results in the expression

$$t=\tau\left\{\ln x+\frac{l^2}{2}\frac{u}{u+1}\frac{1}{ux^2+1}+\right.$$
$$\left(\frac{l^{12}}{2}-\frac{v^4}{2u^2}+\frac{v^4}{u^3}-\frac{2v^3}{u^3}-\frac{1}{2}\right)\ln\left(ux^2+1\right) \tag{20}$$
$$\left.-\frac{v^4}{2u^2}x^2+l^{12}\left[\ln\left(1-x^2\right)^{1/2}\right]\right\}_{x=\cos(\lambda_2-\lambda_1)}^{x=\cos(\lambda_2-\lambda)},$$

where

$$\tau=\frac{\gamma_1 a^2}{3Kl^6(l^2-1)}, \tag{21}$$

$u=l^2-1$ and $v=l^4-1$. Unlike model A, this model does not yield a simple expression for the relaxation time.

3. Experiment and results

3.1. *Sample preparation and apparatus*

The PDLC samples used in this experiment were prepared by phase separation methods using PMMA mixed with the nematic material E7 (EM Chemicals) in a ratio of 1:2 by weight. Acetone was used as a solvent to form a homogeneous mixture and the sample was prepared on a hot plate. A relatively long cooling time was needed to obtain droplets about 1.0 μm in size. The index match resulting from these materials is quite good. Samples of elongated droplets were first prepared between glass substrates with 17.5 μm spacers and with transparent conductive coatings, then warmed to temperatures near the melting point of the polymer and the substrates slightly sheared as illustrated in figure 3 (*a*). It was often observed that there were regions of the sample where droplet alignment was more uniform than in others, but uniformly aligned samples were possible.

Polarized laser light is incident on the sample at an angle θ defined in figures 3 (*b*) and 2 (*b*). A D.C. voltage pulse 2 ms in duration was applied to the indium–tin oxide electrode of the sample and the transmitted light collected in a small collection cone angle of $<2^\circ$. The sample was placed in a cylindrical container with a fluid index matched to the glass to avoid Fresnel corrections. Since the refractive index of the polymer, n_p, is a close match to the index, $n_\perp$, of the liquid crystal, the angle of maximum transmission allowed us to monitor the orientation of **n**, **N** and **a** [14] (see figure 2). Since the collection angle of the detector is small, the measured transmitted light intensity I is related to the total scattering cross-section of the nematic droplets, σ, by the expression $I_0\exp(-\sigma\alpha d)$, where α is the number density of the scatterers, d is the distance the light traverses the sample, $d_0/\cos\theta$, and I_0 is the incident intensity [15, 16].

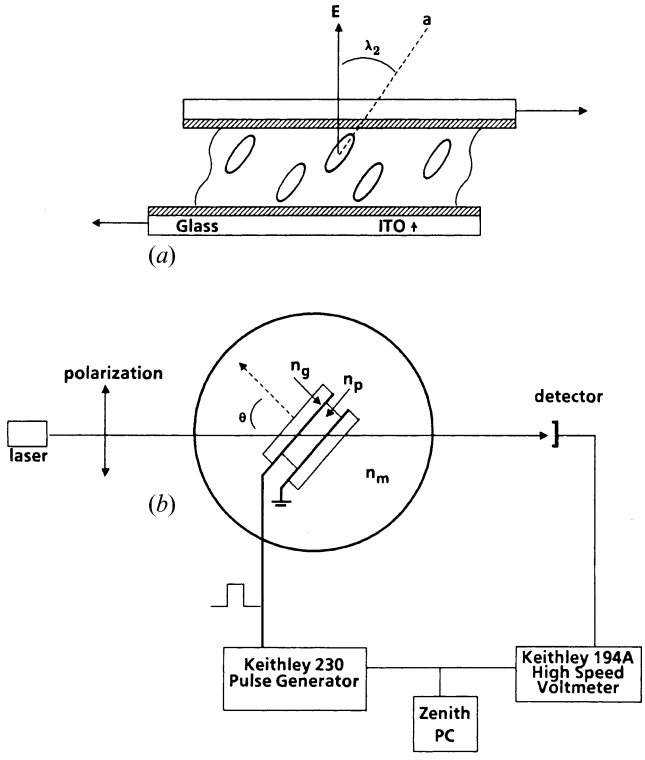

Figure 3. Illustration of (*a*) PDLC sample with aligned and shaped droplets obtained by shearing and (*b*) experimental configuration where the collection angle of the detector is $\leqslant 2^\circ$. The refractive index of the glass substrate, n_g, is approximately matched to that of the fluid in the cylindrical vial, n_m. ITO, indium–tin oxide.

Figure 4 shows the relative transmitted light intensity as a function of the angle of incidence, θ, in the presence and absence of a large saturating voltage applied to the cell. The peak at 66° establishes the orientation of **a** in the sample and the angle λ_2. A single peak in the transmitted intensity at 66° indicates that the refractive

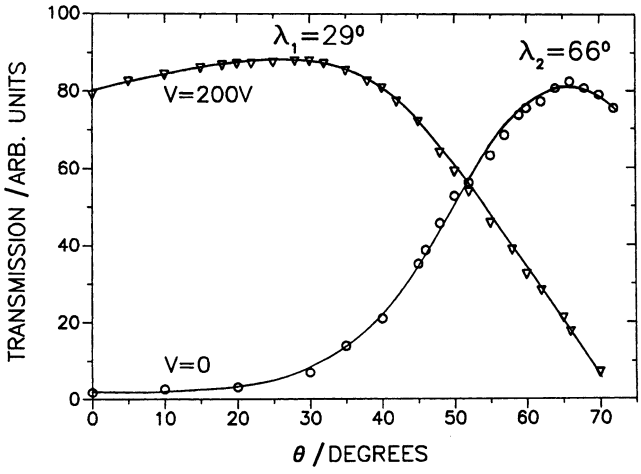

Figure 4. Plot of the relative transmitted intensity versus the angle of incidence for a sheared sample of PMMA–E7 (1:2) with no voltage and with a saturating voltage of 200 V applied to the sample cell.

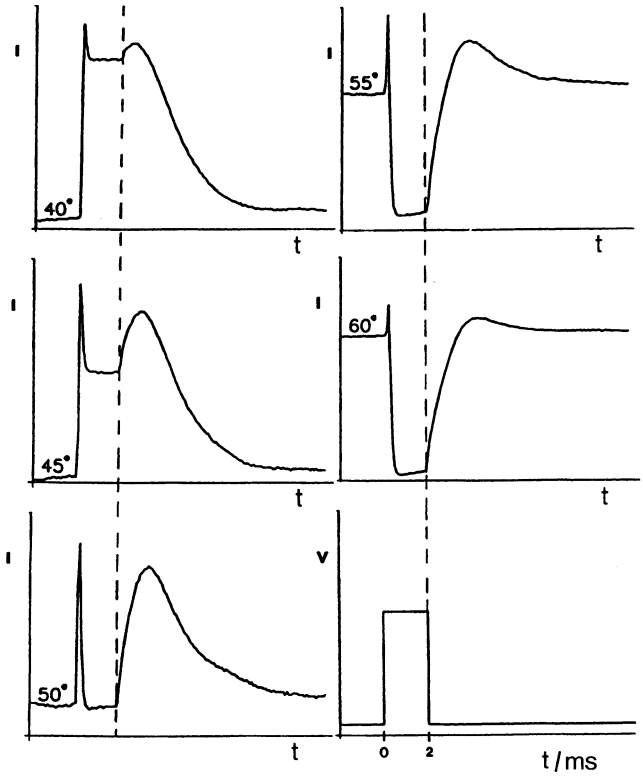

Figure 5. Recordings of the relative transmitted polarized light intensity through a PMMA–E7 sample cell during and following an applied 200 V D.C. 2 ms pulse at selected angles of incidence, θ.

index of the polymer, n_p, is closely matched to $n_\perp$ of the nematic droplet [14]. The breadth of the peak at $\theta=66^\circ$ reflects the distribution in the droplet directors in their equilibrium orientation in the absence of the field. In the presence of the aligning field, the broader peak is due to a broader distribution of droplet directors caused by a non-uniformity in droplet shapes and sizes. The peak at 29° gives the direction of **N** and the angle λ_1. Small variations in l and a distribute the value of λ_1 and hence broaden the peak.

3.2. *Relaxation of the droplet director*

Figure 5 shows typical traces of light transmission resulting from a D.C. pulse 2.0 ms in duration. Representative transmission responses are shown for selected angles, θ, of incident polarized light. The peak in the transmitted intensity following the leading edge of the applied D.C. pulse results when the orientation of the droplet director is parallel to incident beam (index matched orientation) as the nematic droplets respond to the applied field. Following the removal of the D.C. voltage, the transmitted intensity again passes through a maximum as the droplet director relaxes to its equilibrium orientation. The time associated with this second maxima allows us to determine the orientation of **n**, given by the angle λ, as a function of time.

The droplet director angle $\lambda(t)$ is plotted in figure 6 for the relaxing PMMA–E7 sample following the removal of the D.C. pulse. The full curve in the plot is calculated from equation (20) for the ellipsoidally shaped droplet (model B) using parameters listed in the caption of figure 6. For comparison, the broken curve is the best fit to equation (11) of model A. In each case the fitting parameter is the value of l which is $l=1.19$ for model A and $l=1.625$ for model B. It is seen that the ellipsoidally shaped droplet gives a slightly better fit. It appears that there is a large discrepancy in values for the aspect ratio of the droplets between models A and B; however, this can be reconciled by considering the dependence of the radius of curvature of the two models on the aspect ratio. A close inspection of equations (7) and (17) reveals that the shape of model A can be approximated as an ellipse with an aspect ratio of l^2, in which case $(1.19)^2=1.42$ is closer to the valued obtained for l of model B. A comparison of the two models is illustrated in figure 6.

3.3. *Voltage response*

In figure 7 we have plotted the measured values of λ_1 versus the applied voltage for the PMMA–E7 sample. The solid curve is the best fit from model B, where we have used the value of l from the relaxation time measurement. In this case the fit is sensitive to the electronic properties of the polymer and the liquid crystal; the fitting parameters are ratios of the

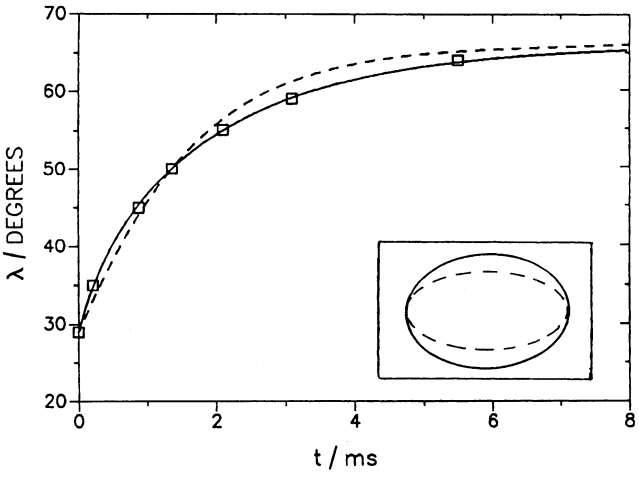

Figure 6. Relaxation of the droplet director, **n**, in terms of the angle λ versus time for the sample of PMMA with E7. The full curve is a fit to equation (20) model B using the parameters $\lambda_1=29°$, $\lambda_2=66°$, $a=0.5\,\mu m$, and $\gamma_1=4\times10^{-2}\,kg\,m^{-1}\,s^{-1}$. The broken curve is a fit to equation (11) of model A with the same values for λ_1, λ_2, γ_1 and a. The fitting parameter in both cases is the aspect ratio l. Model A is best fit with $l=1.19$ and for model B $l=1.625$. The fits indicate that not only the aspect ratio but also the shape of the elongated droplet is significant. The inset shows an ellipse with an aspect ratio of l for the full curve. This is the figure that would result from the radius of curvature described in equation (17) for model B. The broken-curve is an ellipse with an aspect of l^2. This is approximately what would result for the shape of model A with the same length for the major axis of model B.

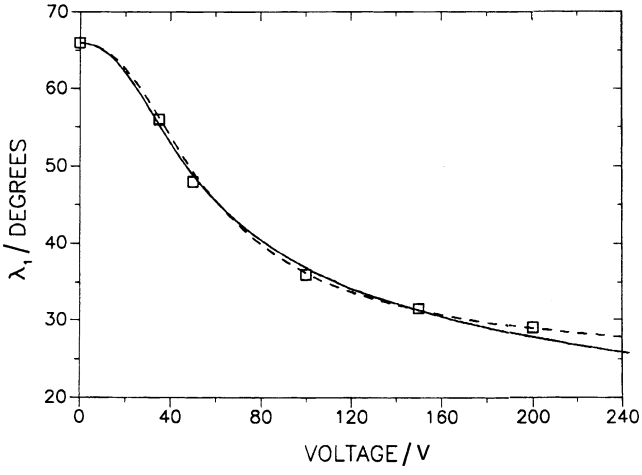

Figure 7. Measured values of λ_1 versus applied voltage for the PMMA–E7 sample. The full curve represents the best fit of equation (19) of model B and the broken curve is the best fit with the same electric field correction as equation (19) but with the smaller radius of curvature from model A. The full curve was fitted with the values $\sigma_\parallel/\sigma_\perp=1.420$ and $\sigma_\parallel/\sigma_1=21.0$. The broken curve was fitted with $\sigma_\parallel/\sigma_\perp=1.510$ and $\sigma_\parallel/\sigma_1=18.0$. The aspect ratios used are those from figure 6 and other parameters include: $d_0=17.5\,\mu m$, $a=0.5\,\mu m$, $\Delta\varepsilon=13.8\ \varepsilon_0$, $\varepsilon_\parallel=19.0\varepsilon_0$, $K=1.71\times10^{-11}\,N$ and $\lambda_2=66°$.

conductivities $\sigma_\parallel/\sigma_\perp$, and $\sigma_\parallel/\sigma_1$. The full curve shows the best fit to equation (19) of model B, with $\sigma_\parallel/\sigma_1=21.0\pm0.5$ and $\sigma_\parallel/\sigma_\perp=1.420\pm0.005$. The broken curve is the best fit using the radius of curvature described in model A, equation (7) where the best fit parameters are $\sigma_\parallel/\sigma_1=18.0\pm0.5$ and $\sigma_\parallel/\sigma_\perp=1.510\pm0.005$.

4. Discussion and conclusions

Experiments have been presented to study the effect of droplet shape on the relaxation time and voltage response of a PDLC light shutter. Models, although admittedly crude, have been presented to quantify the effects of droplet shape on these response characteristics and explain the experimental results. In the case of the relaxation time, the results indicate that the droplet shape can be a dominant feature. Aspect ratios no larger than $l=1.1$ have a significant influence on the relaxation time. This would suggest that droplet shaping, which naturally results from the preparation of PDLC films (without intentional shearing), could be a principal mechanism for relaxation in some, or possibly most, displays, and that the simple equations of model A, (see equations (10), (12) and (16)) may give reasonable estimates for the response of PDLC shutters. In the comparison of the two different droplet shapes, the elliptically shaped droplet model B gave the best fit to the data. This appears to be in agreement with our photographs from scanning electron microscopy of sheared samples (not shown here) which appear to be of an elliptical shape and have aspect ratios of about 1.5–2.0.

The voltage response has been found to be more difficult to understand in that the electric field inside the droplet is substantially reduced from the field V/d_0 applied to the shutter. In the model presented here this reduction is explained in terms of the conductivity ratio between the polymer and liquid crystal, $\sigma_\parallel/\sigma_1$, which had values of approximately 20 for the system studied. This value appears to be small but may be reasonable since the polymer is heavily plasticized with dissolved liquid crystal material and is very soft. The conductivity ratio may, therefore, not be too small. Published result in other thermoplastics have determined that as much as 50 per cent of the liquid crystal remains dissolved while the remainder is phase separated into droplets [14].

We have found that the anisotropy in the conductivity of the liquid crystal was essential to fit Figure 7. The measured ratio of $\sigma_\parallel/\sigma_\perp=1.4$ is in reasonable agreement with that reported in the literature [12].

The model of an isolated spherical droplet used to calculate the internal electric field may be too simplified. It could be that an elongated droplet is also required in this model calculation. The effect of an elongated

droplet tilted with respect to the field would be to change not only the magnitude of the internal field but also its direction. Such calculations are in progress. More experiments on different values of l are intended and it is clear that more-refined theoretical models are necessary. In general, the applied voltage needed to turn the shutter on is proportional to the factor (σ_2/σ_1+2), whereas the turn-on time will be proportional to the square of this factor. Future experiments to obtain independent measurements of $\sigma_{\|}/\sigma_1$ and $\sigma_2(\lambda)$ are under way, as are electro-optic experiments where this ratio is varied to study its effect on the response times and voltages.

The most significant result of this study is that by shaping the droplets we have been able to shorten the relaxation time with little increase in the switching voltage. In recent work with shaped droplets in epoxy materials we have achieved relaxation times of about 5 ms with a switching field of $1\,V\,\mu m^{-1}$.

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