

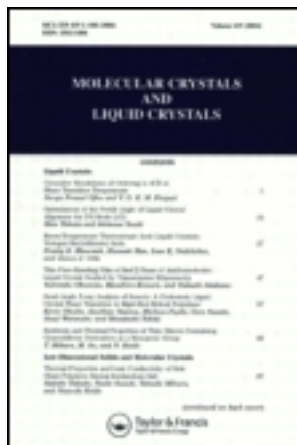
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## Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics

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## Polymer Dispersed Liquid Crystals for Display Application

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# Polymer Dispersed Liquid Crystals for Display Application

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An overview of polymer dispersed liquid crystal (PDLC) materials, their physical properties, and potential applications in the optic and electrooptic industry is presented. These optoelectronic materials have unique properties which are expected to expand liquid crystal technology into new display and light shutter applications. Recent research by small and large industrial and university laboratories on device physics and chemistry has provided substantial progress towards the commercialization of these materials. Work to date on such features as response times, switching voltage, and contrast as well as material preparation procedures and unique optical characteristics will be reviewed. These materials are also of interest to basic physics because of new kinds of physical phenomena brought on by the confinement of a nematic liquid crystal to small submicron-size droplets. Enhanced surface-to-volume ratio, large nematic deformations and problems associated with molecular anchoring at a polymer wall allow for new studies of surface mediated phenomena.

## I. INTRODUCTION

When nematic liquid crystals are dispersed as submicron-size droplets in polymers, electrically switched light shutters with a wide range of applications extending beyond existing liquid crystal technology are possible.<sup>1-5</sup> These applications include large-scale flexible displays that do not require polarizers and are simple and cost effective to fabricate. Switchable coatings for windows to be used for controlling daylight or interior lighting, privacy, cosmetics, solar heat gains, security, etc., provide a totally new application of liquid crystal materials. The absence of polarizers coupled with the electrically con-

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trollable light scattering principle upon which these films operate make them useful for low-loss, high-intensity color projectors, expanding projection technology.<sup>6</sup> Polymer dispersed liquid crystal (PDLC) materials can also be optically switched with high intensity laser light,<sup>7</sup> providing still other types of applications in such areas as optical sensor protection and optical processing and computing. Finally, thermally and mechanically responsive PDLC films make possible thermal and strain sensors with applications in the medical and coatings industries.

A light valve is constructed by sandwiching a 10–25  $\mu\text{m}$  thick film of PDLC material between transparent conducting electrodes.<sup>2</sup> One mode of operation is that illustrated in Figure 1(a) where nematic droplets with a positive dielectric anisotropy are randomly oriented in the film in the opaque state. In this state, the film has a translucent white appearance because of the light scattering properties of the droplets. Upon application of a field the droplets align in a direction parallel to the field. If the refractive index  $n_{\perp}$  (index perpendicular to the nematic director) approximately matches the refractive index of the polymer matrix, the film will switch to a transparent state [Figure 1(b)]. Upon removal of the field, the droplets return to their original random orientation and the film returns to its opaque state. Submillisecond switching times are possible with these films.

The electromagnetic field of a high intensity laser can also be used to switch the films. For example, the electric field of laser light normally incident on the device of Figure 1(b) will compete with the applied field and reorient the droplets to a scattering or opaque state.<sup>7</sup> This is but one example of nonlinear optical devices possible from these films.

Thermally switched films are prepared by matching the refractive index of the polymer with that of the liquid crystal in the isotropic phase,  $n_i$ . Changing the temperature of the film across the isotropic–nematic phase transition reversibly switches the film from opaque to clear as illustrated in Figure 1(d). Mechanically responsive devices operate on the principle that slight modifications in the droplet shape alter the polarization of the scattered light.<sup>8</sup>

Phase separation methods used to prepare PDLC materials are reviewed in Section II. Using these methods, it is possible to make liquid crystal droplets of uniform and predetermined size and shape in order to optimize specific electrooptic properties. The nematic director configuration inside the droplets and methods used to determine that configuration are described in Section III. Electrooptic response of PDLC displays is described in Section IV. The role of index matching and its effect on film transparency and angular dis-

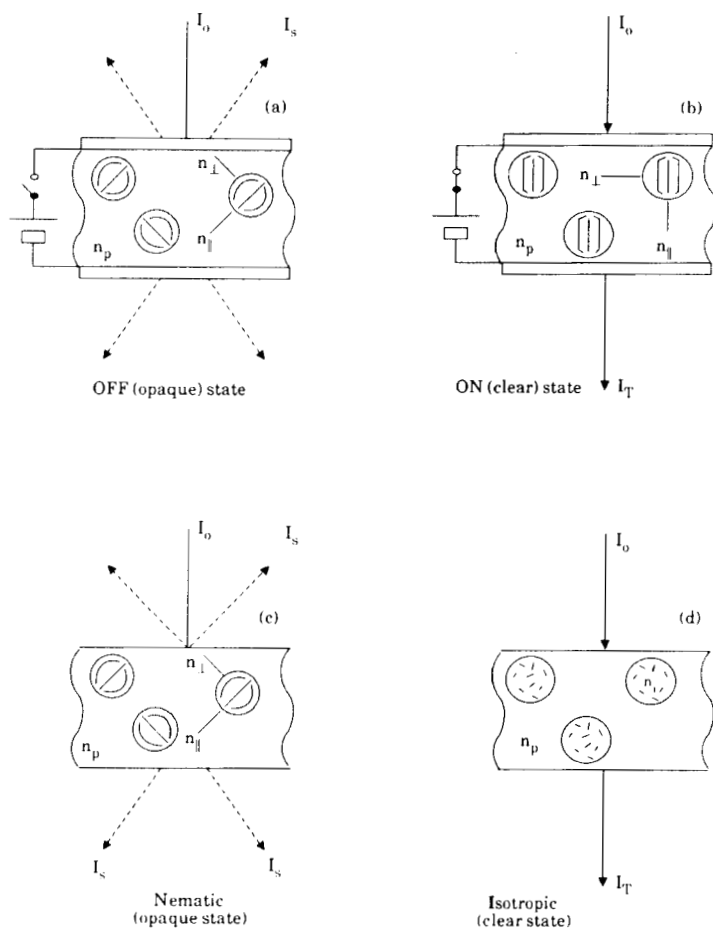


FIGURE 1 PDLC light shutter illustrating the opaque state (a) and (c), the electrooptically switched transparent state, (b), with aligned droplets and the thermally switched transparent state, (d), with the droplets in the isotropic phase.

crimination is discussed in Section V, as are the light-scattering properties of the films and their contrast in display applications. Section VI contains the conclusions.

## II. MATERIALS AND PHASE SEPARATION METHODS

The formation of uniform liquid crystal droplets in a polymer binder may be achieved by several different phase separation processes<sup>2,9</sup> which include phase separation by polymerization, phase separation

by thermal processes, and phase separation by solvent evaporation. Each approach involves forming a homogeneous solution of polymer or prepolymer with a liquid crystal material followed by phase separation, droplet formation, and finally polymer gelation or solidification. These processes are a result of droplet nucleation and growth, and spinodal decomposition.<sup>10</sup> Phase separation processes offer distinct advantages over other suggested means of preparing similar films, such as microencapsulation<sup>3</sup> or use of microporous filters.<sup>1</sup> These advantages include the ability to form droplets of uniform size and to adjust that size over a broad range of droplet diameters extending from 20  $\mu\text{m}$  or larger to as small as 0.01  $\mu\text{m}$ . Phase separation procedures can be applied to a broad range of polymers including thermoplastics,<sup>9</sup> thermoset polymers,<sup>2</sup> or uv-cured systems.<sup>4</sup> Phase separation methods are normally simple, one-step processes which can reduce fabrication costs in display and window applications.

#### **A. Phase separation by polymerization**

Phase separation by polymerization is useful when prepolymer materials are miscible with low molecular weight liquid crystal compounds. A homogeneous solution is made by mixing the prepolymer with the liquid crystal. Polymerization is achieved through a condensation reaction, as with epoxy resins<sup>2</sup>; through free radical polymerization, as with a vinyl monomer catalyzed with a free radical initiator such as a benzoylperoxide; or through a photo-initiated polymerization. The solubility of the liquid crystal decreases in the lengthening polymers until the liquid crystal phase separates, forming droplets. The droplets grow until gelation of the polymer locks in the droplet morphology. Droplet size and morphology are determined during the time between droplet nucleation and gelation of the polymer. Size is controlled by the rate of polymerization, the relative concentrations of materials, the types of liquid crystal and polymers used, and by such physical parameters as viscosity, rate of diffusion, and solubility of the liquid crystal in the polymer. The rate of polymerization is controlled by cure temperature for thermally cured polymers or by light intensity for photochemical polymerization.<sup>4</sup>

Droplets of uniform size can be achieved as is illustrated by the electron microscope photograph of Figure 2. One method of controlling droplet size in some materials is to control the rate of polymerization by the temperature of cure. Figure 3 shows the effect of cure temperature on droplet size and density in an epoxy system. Transmission electron microscopy and nuclear magnetic resonance

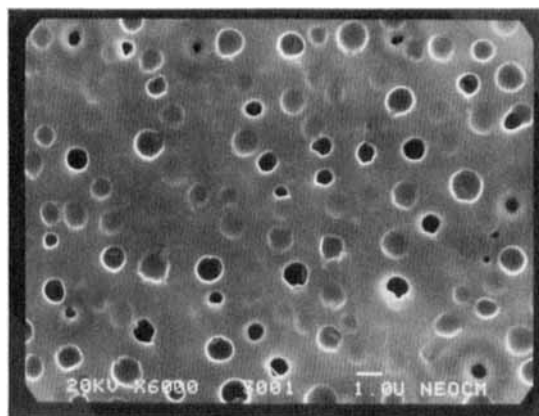


FIGURE 2 A scanning electron microscope photograph of a PDLC material formed by polymerization induced phase separation using Epon/capcure/E7 mixture of epoxy/curing agent/liquid crystal in 1:1:1 proportion cured at 60°. The size of the droplets is 1.0  $\mu\text{m}$  in diameter.

have confirmed that we could prepare materials with droplet sizes less than 0.01  $\mu\text{m}$ .<sup>11</sup> Droplets of this size are useful in studying the effects of a surface on nematic liquid crystalline order.<sup>11</sup> In display applications diameters in the range of 0.3 and 3.0  $\mu\text{m}$  are normally desirable.

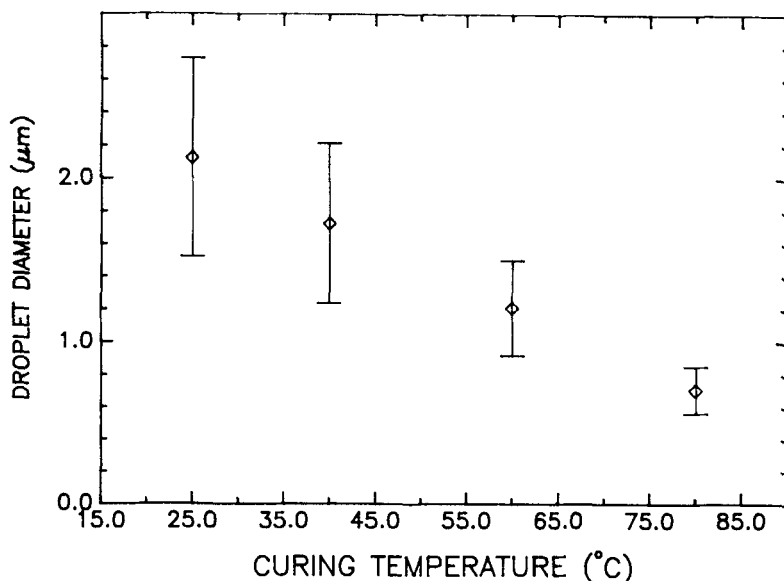


FIGURE 3 Droplet size versus cure temperature of an Epon/capcure/E7 mixture.

## B. Phase separation by thermal processes

Phase separation by thermal processes is useful for thermoplastics which melt below their decomposition temperature. This process is illustrated in Figure 4(a). A binary mixture of polymer and liquid crystal forms a homogeneous solution at elevated temperature. Cooling the homogeneous solution into the miscibility gap along line *A–B* in Figure 4(a) causes phase separation of the liquid crystal. The droplet size of the occluded material is governed by the rate of cooling and depends upon a number of material parameters, which include viscosity, chemical potentials, etc. Polymers such as polymethylmethacrylate (PMMA) and polyvinylformal (PVF) form droplet dispersions with cyanobiphenyl liquid crystal materials.<sup>12</sup> In general, larger concentrations of liquid crystals are required for these films as compared to phase separation by polymerization. Aligned droplets can be made with some thermoplastic materials if a field is applied during the droplet formation process. After the droplets are formed and the field removed, droplets with bipolar alignment have been found to remain oriented.<sup>13</sup> This offers one form of thermal memory for use in irreversible thermometers.

## C. Phase separation by solvent evaporation

Phase separation by solvent evaporation is useful with thermoplastics which melt above the decomposition temperature of the thermoplastic or the liquid crystal, or where solvent coating techniques are used. The liquid crystal and polymer are dissolved in a common solvent forming a homogeneous solution. The solvent is then removed by evaporation, resulting in phase separation and polymer solidification. This process may be understood through the ternary phase diagram illustrated in Figure 4(b). A homogeneous solution of composition *A* is made. Solvent evaporation moves the system along line *A–B*. Passage across the miscibility gap results in droplet formation and growth until polymer solidification locks in the droplet morphology. The final material at point *B* will have liquid crystal droplets dispersed in a polymer binder, with a droplet size dependent on the rate of solvent removal. The polymer will be plasticized by dissolved liquid crystal which will lower the glass transition temperature of the polymer and enhance its moldability and processability.

Solvent evaporation can be used in conjunction with thermal processing for materials that melt below the decomposition temperature. Films are formed on a suitable substrate using standard film coating techniques such as web coating or doctor blading. The solvent is



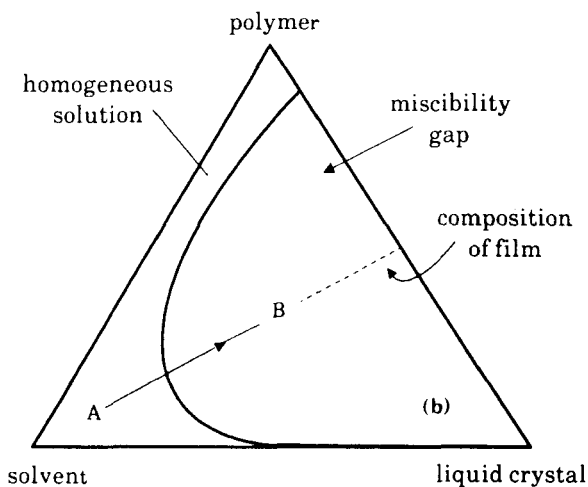
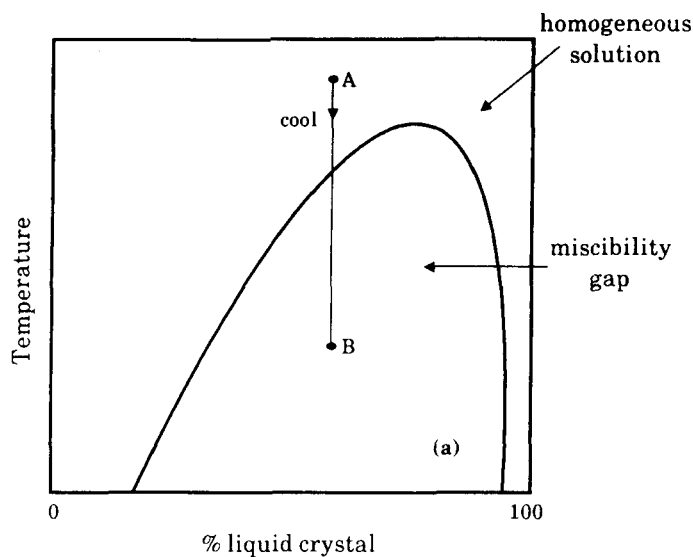


FIGURE 4 Schematic equilibrium diagram of a binary mixture of liquid crystal compound and polymer illustrating droplet formation by thermally induced phase separation, (a); and a ternary mixture of solvent, polymer, and liquid crystal illustrating droplet formation from phase separation induced by solvent evaporation, (b).

rapidly removed with no concern of droplet size or density. Further processing, such as laminating a cover substrate, is performed followed by a final annealing step, where the film is warmed to redissolve the liquid crystal in the polymer and cooled at a rate chosen to give the desired droplet shape and density. This combination of the solvent evaporation process followed by thermal annealing adds flexibility and ease to the production of commercial PDLC films.

### III. DIRECTOR CONFIGURATIONS

An important feature in understanding the physics of display applications is the director configuration of a nematic liquid crystal when confined to a polymer cavity. Many different configurations are possible and they react differently to an applied field. The configuration of the nematic director depends on how the molecules are anchored at the droplet wall, droplet shape and size, material parameters such as the deformation constant of the liquid crystal, and strength of the applied field.

In this paper we review a few simple director configurations calculated in the approximation of a single elastic constant  $K_1 = K_2 = K_3 = K$ . In this case the external field and the elastic contributions to the free energy of a nematic droplet can be written as<sup>8</sup>

$$F = \frac{1}{2} \int \left[ K[(\nabla \cdot \mathbf{n})^2 + (\nabla \times \mathbf{n})^2] - \epsilon_0 \Delta \epsilon (\mathbf{E} \cdot \mathbf{n})^2 \right] dV \quad (1)$$

where  $\Delta \epsilon$  is the dielectric anisotropy ( $\epsilon_{\parallel} - \epsilon_{\perp}$ ). We neglect surface-induced effects on the value and anisotropy of the order parameter<sup>8,14</sup> and assume strong molecular anchoring on the droplet surface. The minimization of the free energy  $F$  of Equation (1) results in a partial differential equation<sup>8</sup> which is solved using the nonlinear over-relaxation method.<sup>15</sup> Since tangential wall anchoring tends to be the most prevalent for droplets in the polymers that we have studied we show the bipolar configuration in Figure 5(a) calculated from Equation (1). This is only one structure which can result from tangential anchoring. Williams<sup>16</sup> has predicted a twisted structure which can also result from tangential wall anchoring for certain ratios of the deformation constants and a toroidal-type structure has recently been observed in large droplets by Drzaic.<sup>17</sup>

A weak electric field easily aligns the symmetry axis of a spherical droplet in a direction parallel to the field for the case of a positive

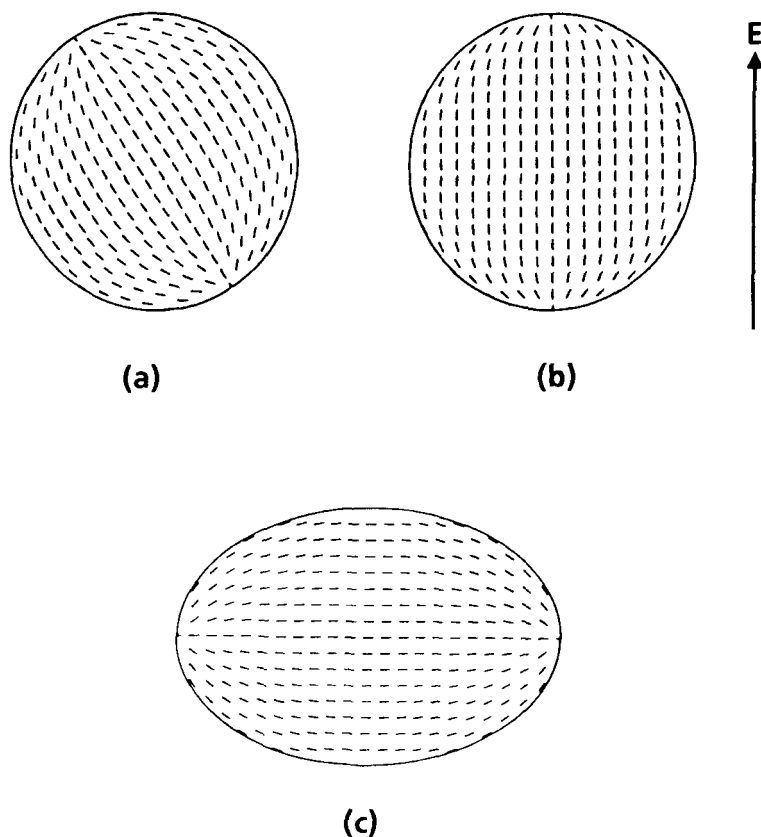


FIGURE 5 The bipolar configuration obtained from tangential anchoring in the absence of a field, (a); in the presence of a strong electric field, (b); and in an elongated cavity, (c). Three-dimensional patterns can be obtained by rotation of these figures about the symmetry axis.

dielectric anisotropy. Strong fields distort the director configuration inside the droplet but only very little in the case of the bipolar configuration, as illustrated in Figure 5(b), a feature which is most important in determining response time in some PDLC materials. We shall show later that in the case of the bipolar configuration the shape of the polymer cavity is important in the relaxation time of a display. The bipolar configuration in an elongated shaped cavity is shown in Figure 5(c). Orientation of the bipolar configuration parallel to the long axis of the cavity minimizes both the bend and twist deformation energies. Such elongated cavities are experimentally achieved by uniaxial mechanical stress. Differences in thermal expansivities of the

polymer and display cell substrate can also cause a stress to create shaped droplets.

A computer simulation for the case of normal anchoring is calculated from Equation (1) and illustrated in Figure 6(a). In this case there is a large distortion in the presence of an applied field, as illustrated in Figure 6(b). In this case the director configuration strongly depends on the strength of the applied field. For  $R/\xi < 4$ , where  $R$  is the radius of a spherical droplet, and  $\xi = (K/\epsilon_0 \Delta\epsilon)^{1/2}/E$  is the coherence length, the lowest free energy corresponds to the "star" configuration, with a point defect in the center. At a value of  $R/\xi \approx 4$ , there is a transition to a configuration with an equatorial line disclination characteristic for  $R/\xi > 4$  [see Figure 6(b)]. The response of a display cell with the star configuration would therefore be expected to be quite different from that of the bipolar structure.

Director configurations in the case of strong anchoring in large spherically shaped droplets ( $\geq 10 \mu\text{m}$ ) have been investigated by several authors.<sup>18-20</sup> For such large droplets, however, the director configurations can be directly observed with an optical microscope and there appear to be a large number of other possible configurations. Recently Yang *et al.*<sup>21</sup> have extended the range of possibilities which are strongly dependent on the coupling constant of the surface potential. They find the director configuration to be dependent on droplet size. Allender and Žumer<sup>22</sup> have further extended the early calculations of director configurations to include spatial variations of molecular order in the droplet; Golemme *et al.*<sup>13</sup> have studied the effects of an applied field and of droplet shape on the director configuration.

It is necessary to note that by using Equation (1) we have ignored variations in the local dielectric anisotropy due to the director con-

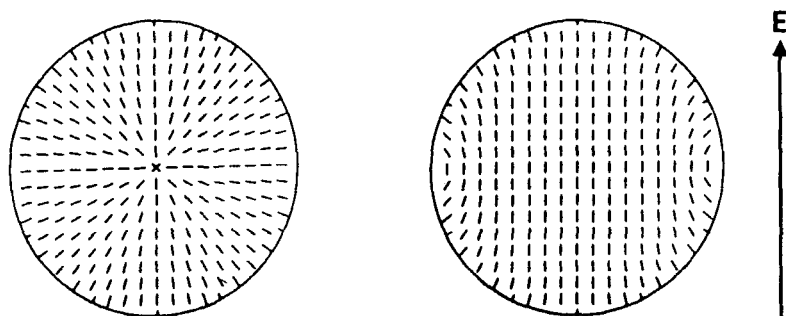


FIGURE 6 The radial configuration obtained from perpendicular anchoring in the absence of a field, (a); and in the presence of a strong field, (b).

figurations in the droplet. Risser et al.<sup>23</sup> have shown this variation in anisotropy to have a large effect on the director configuration in the presence of an applied field. They have explored its effect on several different configurations.

The most effective means we have found to date of observing the director configuration in submicron-size droplets is by deuterium NMR.<sup>13</sup> The deuterium spectrum from a liquid crystal compound deuterated at a particular site is directly related to the director configuration in the droplet. NMR has also been used to study the effect of droplet confinement on orientational ordering.<sup>24</sup> These effects are clearly evident at temperatures near the nematic–isotropic, N–I, transition. The temperature of the transition is shifted and for small enough droplets,  $R < 0.03 \mu\text{m}$ , the first order N–I transition is replaced by a continuous evolution of order. A paranematic order<sup>25</sup> replaces the isotropic order in this size droplet.

#### IV. ELECTROOPTIC RESPONSE

The response times of PDLC shutters are governed by a number of factors: the size, shape and director configuration of the nematic droplet; the value of the deformation constants and rotational viscosity coefficient; the strength of molecular anchoring at the droplet wall. As described in the previous section, the bipolar configuration is dominant in most PDLCs studied to date. 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 5(b). For a perfectly spherically shaped polymer cavity, no 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 as to 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.

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 energy

density inside the droplet is governed by the radius of curvature of the cavity.<sup>26</sup> It was shown in the previous section that in a droplet which is slightly elongated in shape the bipolar axis will prefer to orient along the long axis of the cavity [Figure 5(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 energy density is  $K_{33}/2R^2$  where  $K_{33}$  is the bend constant and  $R$  the radius of curvature at the equator.

We have generalized this concept to model the energy density,  $F_d$ , of a nematic droplet in an elongated cavity, following the expression  $F_d = K/2r^2$  where  $r$  is the radius of curvature along the direction  $\mathbf{r}$ , illustrated in Figure 7, and  $K$  is some effective deformation constant.<sup>26</sup> Ignoring the details of the director configuration, we assign a droplet director 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. For the shape of the elongated cavity we take the radius of curvature following the equation:

$$r = a \left[ \cos^2 \theta + l^2 \sin^2 \theta \right]^{-1/2} \quad (2)$$

where  $l = a/b$  and  $a$ ,  $b$  and  $\theta$  are defined in Figure 7. In this case

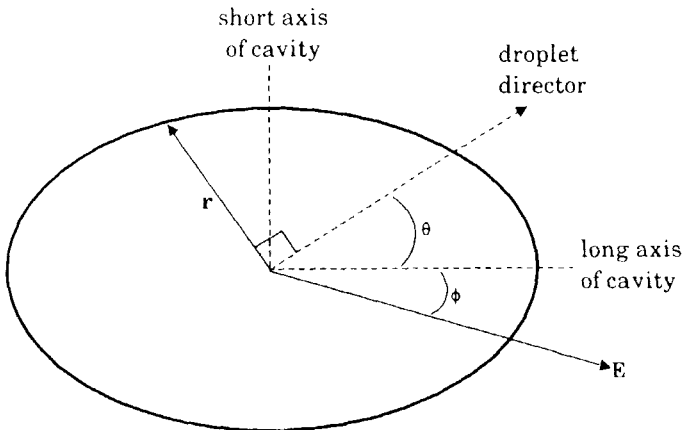


FIGURE 7 Illustration of the droplet director arbitrarily oriented in an elongated cavity where  $\mathbf{r}$  is position vector (perpendicular to the director) indicating the point on the walls where the radius of curvature is of interest.

the elastic torque,  $\Gamma_d = dF_d/d\theta$ , becomes

$$\Gamma_d = \frac{-K(l^2 - 1)}{2a^2} \sin 2\theta \quad (3)$$

The electric torque,  $\Gamma_e$ , is given in its usual form as<sup>27</sup>:

$$\Gamma_e = \frac{\epsilon_0 \Delta \epsilon}{2} E^2 \sin 2(\phi - \theta) \quad (4)$$

where  $E$  is the electric field at the site of the nematic droplet,  $\Delta \epsilon$  is the dielectric anisotropy, and  $\phi$  the orientation of the elongated cavity relative to the direction of the electric field. The viscous torque,  $\Gamma_v$ , is given by<sup>27</sup>:

$$\Gamma_v = \gamma_1 d\theta/dt \quad (5)$$

where  $\gamma_1$  is the rotational viscosity coefficient. Flow inside the droplet has been neglected.

#### A. Relaxation time

The relaxation time of a shutter upon removal of the applied field is calculated by setting  $\Gamma_d + \Gamma_v = 0$  whereby one obtains a characteristic time,  $\tau_{\text{off}}$ , given by the equation<sup>26</sup>:

$$\tau_{\text{off}} = \frac{\gamma_1 a^2}{K(l^2 - 1)} \quad (6)$$

for an elongated droplet of aspect ratio  $l$ . For a slightly shaped droplet of  $l = 1.1$ , a droplet size of  $a = 0.7 \mu\text{m}$  and material constants of  $K = 1 \times 10^{-11} \text{ N}$  and  $\gamma_1 = 5 \times 10^{-2} \text{ kg/ms}^2$ , we obtain a value of  $\tau_{\text{off}} \approx 10 \text{ ms}$  which is not atypical of a PDLC shutter. The squared dependences of  $l$  and  $a$  illustrate the dominant effect droplet shape and size can have. This model and variations of it have been studied by electrooptic measurements on droplets of uniform shape and alignment in specifically designed experiments and were found to be in reasonable accord with the results.<sup>26</sup> Submillisecond switching is readily achieved by reducing the droplet size or by strongly shaping the droplets. Figure 8 shows the temperature dependence of the relaxation for the liquid crystal material E7 in an epoxy binder.

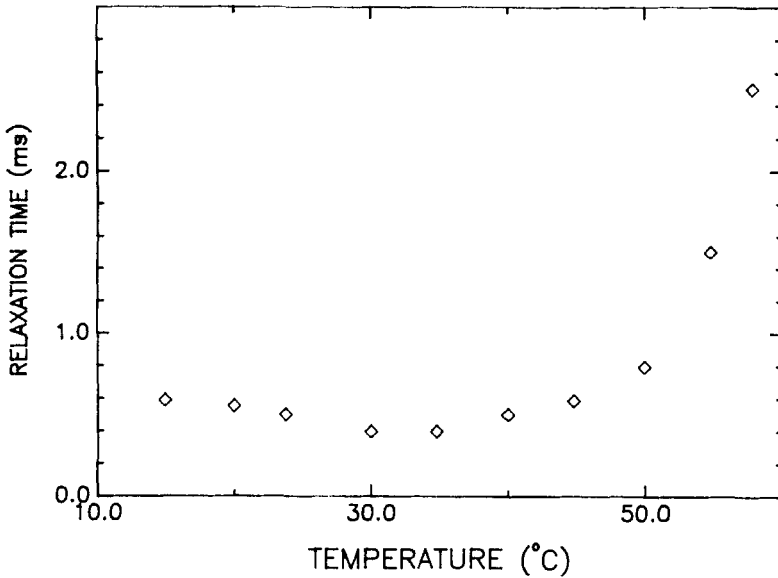


FIGURE 8 Temperature dependence of the relaxation time in an epoxy-based PDLC system with droplets of size  $0.4\ \mu\text{m}$  in diameter. The shape of the droplets were not characterized.

### B. Response time and voltage

The model will give a well defined threshold for the switching voltage when the droplet cavity is oriented perpendicular to the direction of the applied field. This threshold,  $E_{\text{th}}$ , has been found to be<sup>26</sup>:

$$E_{\text{th}} = \frac{1}{a} \left[ \frac{K(l^2 - 1)}{\epsilon_0 \Delta\epsilon} \right]^{1/2}. \quad (7)$$

Using earlier values for  $a$ ,  $K$ , and  $l$  and a value of  $\Delta\epsilon = 14\epsilon_0$ , we obtain a value of  $E = 0.2\ \text{volt}/\mu\text{m}$ . We must recognize, however, that in Equation (7)  $E$  is the electric field experienced by the droplet and can be substantially reduced from the value of  $V/d_o$  where  $V$  is the voltage applied to the cell and  $d_o$  is the cell thickness. This reduction can be caused by several effects, including the resistivity and dielectric constants of the polymer and liquid crystal. In fact, the local field inside the droplet can be spatially dependent due to director deformation.<sup>23</sup>



The response time,  $\tau_{\text{on}}$  for the shutter to turn ON has also been calculated from this model to be<sup>26</sup>:

$$\tau_{\text{on}} = \frac{1}{\gamma_1} \left[ \epsilon_0 \Delta \epsilon E^2 - \frac{K(l^2 - 1)}{a^2} \right]. \quad (8)$$

For large voltages applied to the shutter,  $\tau_{\text{on}} = \Delta \epsilon E^2 / \gamma_1$ . Since the ON state is driven by the field,  $\tau_{\text{on}}$  is normally shorter than  $\tau_{\text{off}}$ .

We have explored other models for droplet shape, such as the ellipsoid of revolution.<sup>26</sup> This shape provides a larger radius of curvature than the model above and gives a stronger dependence of  $\tau_{\text{off}}$  on the aspect ratio,  $l$ , of the droplet. More refined models on the effect of droplet shape have yet to be worked out. Finally we point out that we have only considered here the response of a single droplet. A more complete theory must take into account a collection of randomly ordered droplets and the optical response must include the effect of light scattered by the droplets.

For droplet configurations such as the radial one of Figure 6, factors other than shape may be dominant. In the radial configuration there is a critical field  $E_c$  at which the droplet will switch from the radial to the partially aligned configuration. This provides for different response equations than those of Equations (6)–(8). As described in the previous section the threshold field,  $E_c$ , at which the droplet changes its configuration is  $R/\xi = 4$  whereby:

$$E_c \approx \frac{4}{R} \left( \frac{K}{\epsilon_0 \Delta \epsilon} \right)^{1/2}. \quad (9)$$

Using a cyanobiphenyl compound, a droplet of radius  $1.0 \mu\text{m}$  requires a driving field of  $\sim 4.0 \text{ volt}/\mu\text{m}$  which is substantially larger than that of the bipolar droplet. As with the case of the bipolar droplet, the value of  $E_c$  cannot be directly related to the applied voltage without taking account of the electronic properties of the polymer and liquid crystal. In general  $E < V/d_o$ .

The voltage response for an index matched epoxy is shown in Figure 9. The driving power depends on the resistivity,  $\rho$ , of the polymer. Polymers, such as PMMA, have a high resistivity and the driving power is  $\leq 1.0 \mu\text{W}/\text{cm}^2$ . Epoxy materials generally require a higher driving power,  $\sim 1.0 \text{ mW}/\text{cm}^2$ . Advantage can be taken of the high resistivity of some PDLC shutters to provide a form memory. The

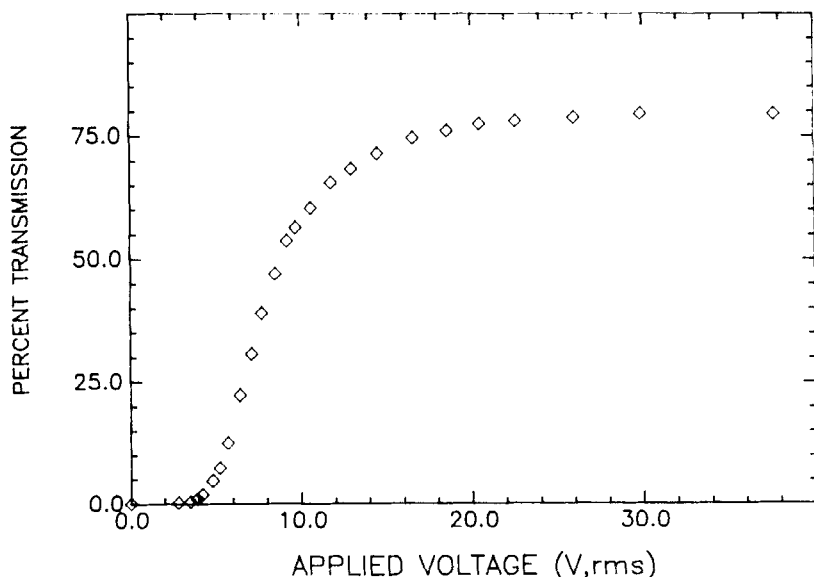


FIGURE 9 Percent of transmitted light through a PDLC light shutter (uncorrected for surface reflections at the glass–air interface) versus an applied 100 Hz voltage. The PDLC sample is an epoxy system (Epon 828 and MK107) index matched to  $n_1$  of E7 (EM Chemicals).

memory time is the product,  $\rho\epsilon$ , where  $\epsilon$  is the dielectric constant of the material. Values of  $\rho\epsilon > 1.0$  seconds are easily achieved in thermoplastics. The display cell is returned to the opaque state by a short circuit.

If the response of the display is driven by a Freedericksz-type transition, as in the radially configured droplet, the relaxation time does not rely on shaping the droplet. An exact calculation of the relaxation time in this case has not been made but can be estimated by assuming a linear relationship for the elastic torque,  $(2\pi)^2 K\theta/R^2$ , where  $R$  is the droplet radius.<sup>28</sup> In the relaxation mode the elastic torque competes with the viscous torque to give a relaxation time of  $\gamma_1 R^2/(2\pi)^2 K$  which is submillisecond for typical PDLC droplet sizes. PDLC displays made from films with radial configured droplets could provide a sharper voltage threshold and fast relaxation time.

## V. INDEX MATCHING, LIGHT SCATTERING, AND SHUTTER CONTRAST

The optical properties of a PDLC film in its transparent state depend strongly upon the value of the perpendicular refractive index of the

nematic,  $n_{\perp}$ , relative to the isotropic index of the polymer,  $n_p$ . Earlier, we only discussed the index-matched condition,  $n_p/n_{\perp} = 1$ . In this condition, light transmission through a film in an applied field of sufficient strength to align the nematic droplets is maximum at normal incidence to the film's surface. The angular breadth of the transmission peak is also broadest in this matched case.<sup>12</sup> The transmission peak narrows while remaining centered at normal incidence as the ratio  $n_p/n_{\perp}$  becomes less than one.<sup>12</sup> For ratios of  $n_p/n_{\perp} > 1$  the transmission peak is shifted away from normal incidence<sup>12</sup> with the angle of maximum transmission depending upon the value of the ratio  $n_p/n_{\perp}$ .

To explain this feature it is useful to consider an effective refractive index,  $n_{\text{eff}}$ , for a droplet in the presence of an aligning field. For light polarized as indicated by the inset of Figure 10(a) the effective index is given by:

$$n_{\text{eff}} = \frac{\bar{n}_{\perp} \bar{n}_{\parallel}}{(\bar{n}_{\perp}^2 \sin^2 \alpha + \bar{n}_{\parallel}^2 \cos^2 \alpha)^{1/2}} \quad (10)$$

where  $\bar{n}_{\perp}$  and  $\bar{n}_{\parallel}$  are the average refractive indices of the field aligned droplet which depend on its resulting director configuration. It is clear that in the case where  $n_{\parallel} > n_{\perp}$ , we have  $\bar{n}_{\perp} > n_{\perp}$  and  $\bar{n}_{\parallel} < n_{\parallel}$  because, on the average, each component contains small admixtures of the other. For the matched case of  $n_p = n_{\text{eff}} > \bar{n}_{\perp}$  maximum transmission through a film will therefore occur at an angle given by:

$$\alpha = \sin^{-1} \left\{ \left[ \left( \bar{n}_{\perp}/n_p \right)^2 - 1 \right] / \left[ \left( \bar{n}_{\perp}/\bar{n}_{\parallel} \right)^2 - 1 \right] \right\}^{1/2}. \quad (11)$$

Figure 10 shows the angular dependent light transmission through two different PDLC films in the ON state. Figure 10(a) is nearly matched while Figure 10(b) is mismatched. The PDLC films were made from the same epoxy polymer but with a different concentration of liquid crystalline material. The lowest concentration is nearly index matched while the higher concentrations become progressively more mismatched with the angle  $\alpha \approx 30^\circ$  for Figure 10(b). The changing concentration of liquid crystal material affects both  $n_p$  and  $\bar{n}_{\perp}$ . The value of  $n_p$  is affected by dissolved liquid crystal in the polymer binder and  $\bar{n}_{\perp}$  is affected by the droplet size. The droplet diameter becomes larger for larger concentrations of liquid crystal and the larger droplets are better aligned by the field and yield smaller values of  $\bar{n}_{\perp}$  creating a larger mismatch with  $n_p$ .

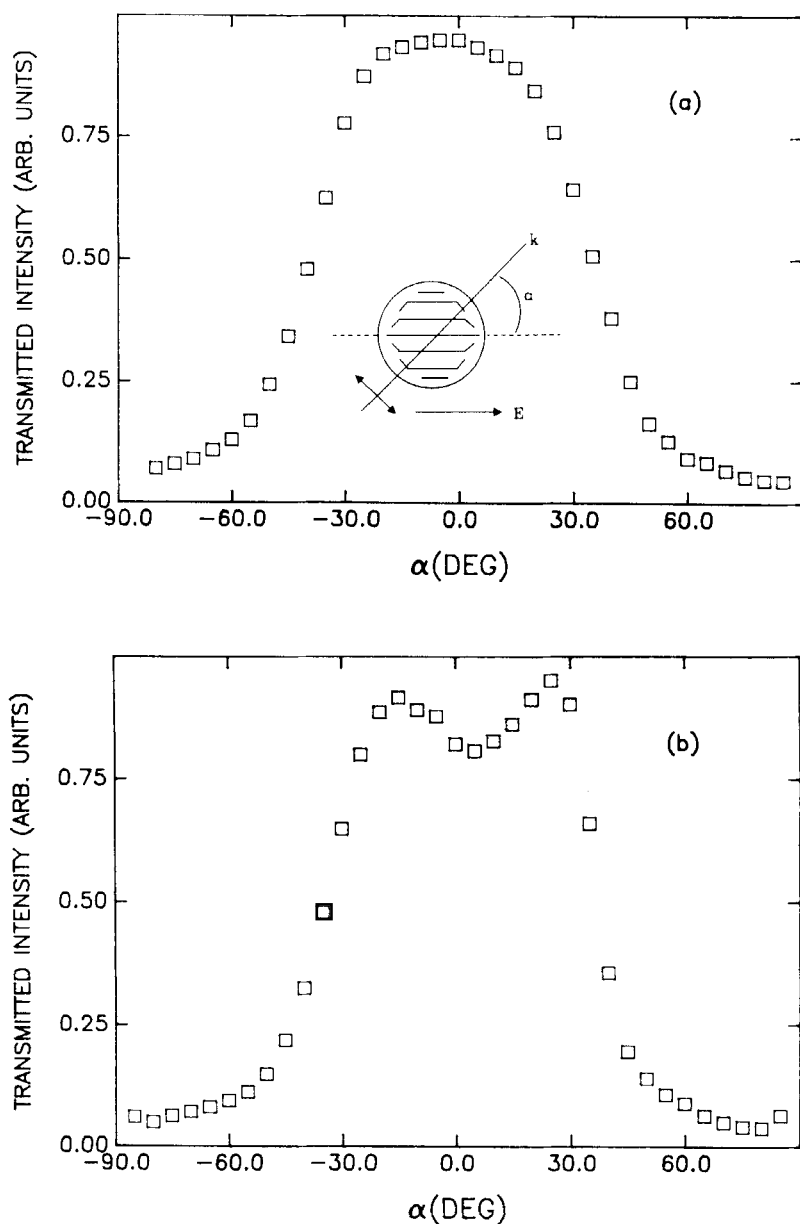


FIGURE 10 Angular dependence of the transmitted intensity for polarized light through PDLC films in the field ON state for two samples of Epon/capcure binder with liquid crystal E7 (EM Chemicals) in relative concentration; 33.1% liquid crystal with an average droplet radius of 0.7  $\mu\text{m}$ , (a); and 37.9% liquid crystal with an average droplet size of 1.2  $\mu\text{m}$ , (b).

Scattering at large angles of incidence causes a haze to the transparent state of a display when lit by diffuse light. Such scattering can cause a reduction in the contrast of the display. Advantage can be taken of the angular dependence for some applications such as light and solar control of switchable window coatings. For a film in its transparent state, transmission coefficients of 85% are achievable at normal incidence wherein light loss from reflection at the air-glass interface of the cell exceeds that from scattering by the film.

In the opaque state most of the light is forward scattered.<sup>8</sup> The relative amount of forward-to-backward scattering depends upon the director configuration in the droplet, the droplet orientation, and the droplet size and shape.<sup>8</sup> The relative amount of back-scattered to forward-scattered light increases as the droplet size is reduced but in practice rarely exceeds 40% and is often only 10–20%.<sup>5</sup>

The theory of light scattering of a nematic droplet has been studied by Žumer.<sup>8,29</sup> He has calculated the differential and total scattering cross section from a single droplet in both the Rayleigh-Gans<sup>8</sup> and anomalous diffraction regime.<sup>29</sup> Experimental measurements by Montgomery<sup>5</sup> indicate that multiple scattering may be important in PDLC films even in their ON state. Multiple scattering is, in part, produced by total internal reflection at the air-sample interfaces for certain scattering angles. Figure 11 shows the differential scattering cross section as a function of angle that we have measured for normally incident polarized light from an index matched PDLC film (the same shutter as used in Figure 9). The shutter was placed in a cylindrical container of glycerol to avoid reflection and refraction problems of a planar air-glass interface. The data near 0° and 90° are masked by residual scattering of the sample container and structure in the plot is clearly evident near 30°.

The contrast in transmission between the ON and OFF states can be quite large for laser light or a well collimated light beam. For collection angles into the detector  $\leq 1.0^\circ$  contrasts  $> 100:1$  are easily achieved between the opaque and transparent states. As the collection angle increases the contrast ratio diminishes because of the collection of more of the scattered light. For projection type displays the collection angle can still be quite small and contrast ratios of 50:1 are achievable. Light shutters are, therefore, quite well suited for low-loss, high intensity projection displays because of this feature and because the transmission is not limited by polarizers and gives a high transparency in the ON state.<sup>6</sup>

When the shutter is used as a display under diffuse lighting conditions, the contrast ratio diminishes. A complete analysis has yet to

be made, but it appears that the haze from the transparent state described earlier contributes to this loss in contrast as well as to the high degree of forward scattering in the opaque state. Nevertheless, high contrast displays are possible particularly with innovative lighting conditions.

## VI. CONCLUSIONS

Major features of light shutters from PDLC films are as follows:

1. They are simple to fabricate by phase separation methods.
2. They do not require polarizers.
3. They are cost effective.
4. They make possible submillisecond switching times.
5. They can be used for large-scale flexible displays and variable transmission window coatings.
6. They operate with low driving power, less than  $1 \mu\text{W}/\text{cm}^2$  achievable with thermoplastics.

At this point of development, it is believed that the best apparent use of these materials is in projection type displays and in large flexible displays for outdoor and indoor use. Switchable windows is a commercial area where these materials may play a unique role in controlling solar heat gains, security, cosmetics, privacy, etc.

PDLC materials can be transformed from a scattering to an absorbing or absorbing/scattering combination by the introduction of dichroic dyes. In this mode of operation the dye must reside primarily in the droplet, phase separating from the polymer. Work on this aspect is currently in progress.

In order to optimize PDLC material for commercial application, more needs to be learned about phase separation processes. It is essential to have good control of droplet size, shape and density. As some director configurations are more desirable than others, more study needs to be done on polymer/liquid crystal surface interactions. Light scattering from PDLC material needs to be better understood. Finally, switching mechanisms require more study, since some polymer binders show different switching behavior.

Rather than being competitive with existing liquid crystal display technology, these materials appear to expand the technology into new areas.

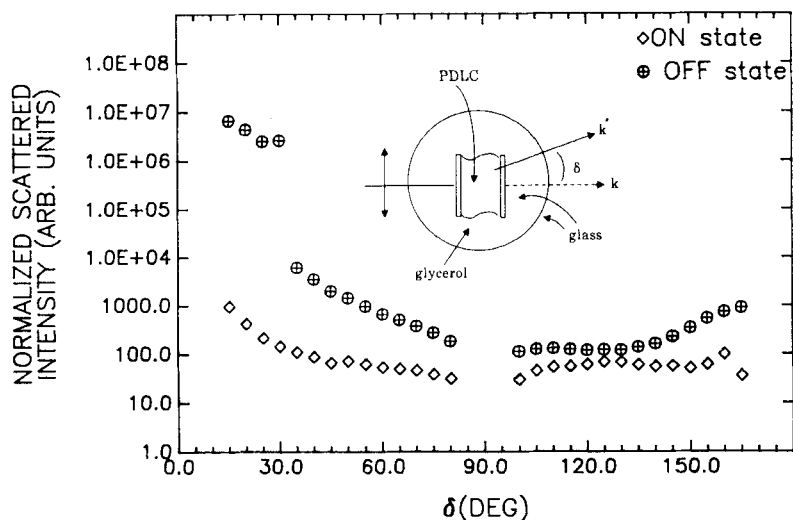


FIGURE 11 Differential scattering cross section measured for a PDLc film from liquid crystal material E7 (EM Chemicals) and an index matched polymer binder from a combination of epoxies Epon 828 and MK107 with capcure as the curing agent. The polarization of the incident light is indicated in the inset as well as the cylindrical sample configuration.

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