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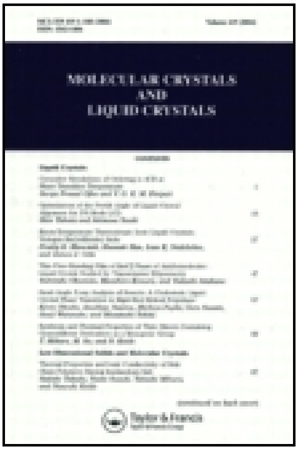

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The Optical Response of Polymer Dispersed Liquid Crystals

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A simple model is proposed to describe the electrooptic response of polymer dispersed liquid crystals. A hierarchy of order parameters is introduced to describe orientational order on different length scales. External fields reorient the liquid crystal in the inclusions, and thus alter the orientational order. The resulting changes in the dielectric and optical properties of the sample are taken into account via changes in the order parameters. The transmittance of PDLC films is considered as a function of static and time dependent magnetic and electric fields.

INTRODUCTION

Polymer dispered liquid crystals are inhomogenous materials, consisting of a low molecular weight liquid crystal dispersed in a polymer.[1,2] These materials may be formed by the phase separation of an initially homogeneus liquid crystal—polymer mixture,[3,4] or by encapsulation of the liquid crystal by the polymer.[5] A wide variety of structures are possible, depending on the material properties of the polymer and the liquid crystal and on their concentration. Since the orientation of the liquid crystal in these structures may be altered by modest fields, they are promising materials for display applications. Polymer dispersed liquid crystal (PDLC) films, consisting of micron sized droplets of a nematic liquid crystal dispersed in a polymer binder, are of particular current interest for projection television, direct view display and switchable windows applications.

Understanding the optical response of these materials is important from both fundamental and applied points of view, and considerable work has been carried out in both measuring and understanding the optical response to electric,[6–10] magnetic[11] and optical[12–15] fields. The structural complexity poses a considerable obstacle to modelling; PDLC films consist of more or less randomly distributed irregular cavities containing the liquid crystal. The heart of the problem is determining the field and liquid crystal configuration everywhere in the film. This is a non-trivial task for liquid crystals even in the simplest geometries, and an exact solution for PDLC films appears prohibitively difficult. In this paper we develop a simple but approximate approach to describe the optical response of PDLC films, based on the assumption that while the applied field reorients the liquid crystal in the inclusions, it does not otherwise significantly change its internal configuration.

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In this model, we introduce a hierarchy of order parameters,[16] and express the optical response in terms of these. Some of the basic elements of the model have been developed previously. The minimization of the free energy of a single ellipsoidal droplet was carried out by Wu *et al.*[6] using a somewhat less general approach than is presented here, although the result is essentially the same. The notion of a droplet director was qualitatively presented by Zumer and Doane.[17] The result for the sample order parameter in the presence of a static magnetic field has appeared before without the details of the derivation.[11]

The utility of the model lies in its ability to describe physical phenomena of interest. Following the development of the sample response, the model is applied to the optical response of PDLC films.

Order Parameters

The anisotropic bulk properties of nematics originate in the orientational order of the molecules. The bulk properties of PDLC material can be related to this orientational order via a hierarchy of order parameters.

If the molecules of the liquid crystal are assumed to possess cylindrical symmetry, where $\hat{\mathbf{l}}$ is a unit vector along the symmetry axis of a molecule, and $\chi_{\parallel}$ and $\chi_{\perp}$ are the molecular diamagnetic susceptibilities parallel and perpendicular to $\hat{\mathbf{l}}$, then the bulk diamagnetic susceptibility can be written, in a uniform homogeneous bulk phase, as

$$\chi_{\alpha\beta} = \rho\frac{(\chi_{\parallel} - 2\chi_{\perp})}{3}\delta_{\alpha\beta} + \rho\frac{2(\chi_{\parallel} - \chi_{\perp})}{3}\left\langle\frac{1}{2}(3l_\alpha l_\beta - \delta_{\alpha\beta})\right\rangle \tag{1}$$

where ρ is the number density and the brackets $\langle\ \rangle$ denote the ensemble average. A suitable order parameter to describe nematic order is of symmetric molecules is therefore

$$Q_{\alpha\beta} = \langle\tfrac{1}{2}(3l_\alpha l_\beta - \delta_{\alpha\beta})\rangle. \tag{2}$$

The symmetric traceless tensor $Q_{\alpha\beta}$ may be diagonalized; if its eigenvectors are $\hat{\mathbf{l}}$, $\hat{\mathbf{m}}$ and $\hat{\mathbf{n}}$, it can be written in terms of the components of these as

$$Q_{\alpha\beta} = \langle\tfrac{1}{2}(3(\hat{\mathbf{n}}\cdot\hat{\mathbf{l}})^2 - 1)\rangle\tfrac{1}{2}(3n_\alpha n_\beta - \delta_{\alpha\beta}) + \tfrac{3}{4}\langle(\hat{\mathbf{l}}\cdot\hat{\mathbf{l}})^2 - (\hat{\mathbf{m}}\cdot\hat{\mathbf{l}})^2\rangle(l_\alpha l_\beta - m_\alpha m_\beta) \tag{3}$$

If the phase is uniaxial, the eigenvalues associated with $\hat{\mathbf{l}}$ and $\hat{\mathbf{m}}$ are identical. In this case, $\langle(\hat{\mathbf{l}}\cdot\hat{\mathbf{l}})^2\rangle = \langle(\hat{\mathbf{m}}\cdot\hat{\mathbf{l}})^2\rangle$, and

$$Q_{\alpha\beta} = S\tfrac{1}{2}(3n_\alpha n_\beta - \delta_{\alpha\beta}) \tag{4}$$

where $\hat{\mathbf{n}}$ is the nematic director and $S = \langle\tfrac{1}{2}(3(\hat{\mathbf{n}}\cdot\hat{\mathbf{l}})^2 - 1)\rangle$ is the scalar order parameter. The physical significance of the director $\hat{\mathbf{n}}$ is that it is parallel to the direction

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of average orientation of the symmetry axes of the molecules, while S gives a measure of the degree of orientational order.

In PDLC samples, although some of the liquid crystal is dissolved in the polymer binder, most of it is contained in micron sized cavities which are distributed throughout the sample volume. In general, both the order parameter S and the director $\hat{\mathbf{n}}$ may vary as a function of position in the nematic liquid crystal contained in the inclusions. We assume that the length scale of spatial variations of the director is much greater than a molecular length, and therefore both $S(\mathbf{r})$ and $\hat{\mathbf{n}}(\mathbf{r})$ can be unambiguously determined at each point in the liquid crystal containing inclusions.

Magnetic Field Induced Order

Field energy and order parameters. The contribution of the interaction of a nematic liquid crystal and a magnetic field $\mathbf{H}$ to the free energy density is

$$\mathcal{F}_H = -\frac{\mu_0}{2}\mathbf{M}\cdot\mathbf{H} = -\frac{1}{2}\mu_0\chi_{\alpha\beta}H_\alpha H_\beta = -\frac{1}{3}\mu_0\Delta\chi H_\alpha H_\beta Q_{\alpha\beta} \tag{5}$$

where $\Delta\chi = \rho(\chi_\parallel - \chi_\perp)$ and we have omitted in the last term the isotropic part of the susceptibility. Explicitly, in terms of the angle between the director and the applied field, the energy density becomes

$$\mathcal{F}_H = -\tfrac{1}{3}\mu_0\Delta\chi H^2 S P_2(\hat{\mathbf{H}}\cdot\hat{\mathbf{n}}) \tag{6}$$

where $P_2(x)$ is the second Legendre polynomial, and $\hat{\mathbf{H}}$ is a unit vector along $\mathbf{H}$. Although most nematic liquid crystals are diamagnetic with $\chi_{\alpha\beta} < 0$, $\Delta\chi$ is positive and these materials prefer to align with the director $\hat{\mathbf{n}}$ parallel to the applied field $\mathbf{H}$. The average magnetic free energy density of the liquid crystal droplet in a uniform applied field $\mathbf{H}$ is

$$\mathcal{F}_H = -\frac{1}{3}\mu_0\Delta\chi H_\alpha H_\beta \frac{1}{V_d}\int_{V_d} Q_{\alpha\beta}(\mathbf{r})\, d^3\mathbf{r} \tag{7}$$

where the integration is over the droplet volume V_d. In analogy with the order parameter for uniform bulk nematics, (cf. Equation 5) it is useful to define an order parameter tensor for the droplet as

$$Q_{d_{\alpha\beta}} = \frac{1}{V_d}\int_{V_d} Q_{\alpha\beta}(\mathbf{r})\, d^3\mathbf{r} = \frac{1}{V_d}\int_{V_d} S\,\frac{1}{2}\,(3n_\alpha(\mathbf{r})n_\beta(\mathbf{r}) - \delta_{\alpha\beta})\, d^3\mathbf{r} \tag{8}$$

The droplet order parameter $Q_{d_{\alpha\beta}}$ is the volume average of $Q_{\alpha\beta}$ over the droplet; it is also symmetric and traceless and may be diagonalized. If $Q_{d_{\alpha\beta}}$ is uniaxial, that is, if two of its eigenvalues are identical, then, as before, it may be written in terms of the eigenvector $\hat{\mathbf{N}}_d$ associated with the unique eigenvalue to give

$$Q_{d_{\alpha\beta}} = \lambda_d \tfrac{1}{2}(3N_{d_\alpha}N_{d_\beta} - \delta_{\alpha\beta}) \tag{9}$$

This defines $\hat{\mathbf{N}}_d$, the droplet director. The associated eigenvalue is $\lambda_d = \langle S\tfrac{1}{2}(3(\hat{\mathbf{N}}_d \cdot \hat{\mathbf{n}}(\mathbf{r}))^2 - 1)\rangle_d$, and the brackets $\langle\ \rangle_d$ now denote the volume average.

In the theory of Oseen and Frank,[18] which is valid in the limit of long wavelength distortions of the director field $\hat{\mathbf{n}}(\mathbf{r})$, the orientational order parameter S is assumed to be constant. For simplicity, we also assume that (with the exception of point and the line defects and in regions very near the surface) S is constant in the droplets. In this case, the eigenvalue λ_d can be written as $\lambda_d = SS_d$ where $S_d = \langle\tfrac{1}{2}(3(\hat{\mathbf{N}}_d \cdot \hat{\mathbf{n}}(\mathbf{r}))^2 - 1)\rangle_d$ is the scalar droplet order parameter. The physical significance of the droplet director $\hat{\mathbf{N}}_d$ is that it is parallel to the direction of average orientation of the nematic director $\hat{\mathbf{n}}$, while the droplet order parameter S_d gives a measure of the degree of orientational order of the nematic director $\hat{\mathbf{n}}$ in the volume V_d. The degree of orientational order of the liquid crystal molecules in the droplet is $\tfrac{1}{2}\langle 3(\hat{\mathbf{N}}_d \cdot \mathbf{l})^2 - 1\rangle_d = SS_d$. Droplet order parameters have been calculated for a variety of director configurations.[7,19] Examples are shown in Figure 1(a).

The free energy per volume of a single uniaxial nematic droplet with constant

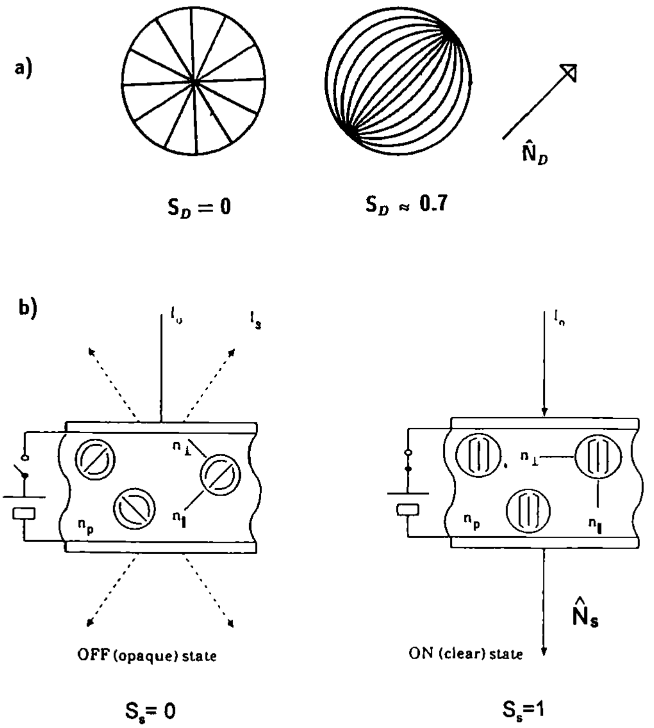

FIGURE 1 Examples of droplet (a) and film (b) order parameters.

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order parameter S and arbitrary director configuration $\hat{\mathbf{n}}(\mathbf{r})$ a uniform magnetic field $\mathbf{H}$ can therefore be written as

$$\mathcal{F}_H = -\tfrac{1}{3}\mu_0\Delta\chi H_\alpha H_\beta Q_{d_{\alpha\beta}} = -\tfrac{1}{3}\mu_0\Delta\chi H^2 S S_d P_2(\hat{\mathbf{H}}\cdot\hat{\mathbf{N}}_d). \tag{10}$$

Since $\Delta\chi > 0$, the liquid crystal prefers to align with the droplet director $\hat{\mathbf{N}}_d$ parallel to the applied field $\mathbf{H}$.

Effects of cavity shape. We next consider the elastic energy associated with distortions of the director field $\hat{\mathbf{n}}(\mathbf{r})$. The Frank free energy density for an arbitrary director field, neglecting surface terms, is given by

$$\mathcal{F}_{el} = \tfrac{1}{2}K_1(\nabla\cdot\hat{\mathbf{n}})^2 + \tfrac{1}{2}K_2(\hat{\mathbf{n}}\cdot\nabla\times\mathbf{n})^2 + \tfrac{1}{2}K_3(\hat{\mathbf{n}}\times\nabla\times\hat{\mathbf{n}})^2 \tag{11}$$

where K_1, K_2 and K_3 are elastic constants associated with splay, twist and bend. In the one constant approximation, where $K_1 = K_2 = K_3 = K$, the free energy density becomes

$$\mathcal{F}_{el} = \frac{1}{2}K\left(\frac{1}{R_1^2} + \frac{1}{R_2^2}\right) \tag{12}$$

where $R_1 = |\nabla\cdot\hat{\mathbf{n}}|^{-1}$ and $R_2 = |\nabla\times\mathbf{n}|^{-1}$ are the radii of curvature of the director field associated with divergence and curl of $\hat{\mathbf{n}}$. In principle, in the absence of external fields, the director configuration of the nematic liquid crystal in the inclusion can be determined by minimizing $\mathcal{F}_{el}$ subject to boundary conditions at the surface.[20-22] In practice, such a calculation is difficult, and we turn therefore to an approximate analytic description.

For simplicity, we assume that the shape of the cavity containing the liquid crystal is an ellipsoid. In terms of a vector $\mathbf{r} = r\hat{\mathbf{r}}$ from its center to a point on its surface, its equation is $R^2 = r_\alpha r_\beta \Lambda_{\alpha\beta}$ where $\Lambda_{\alpha\beta}$ is the characteristic tensor of the cavity shape. For a prolate ellipsoid of revolution, $\Lambda_{\alpha\beta} = (\delta_{\alpha\beta} - \frac{2}{3}\varepsilon^2\frac{1}{2}(3L_\alpha L_\beta - \delta_{\alpha\beta}))$ where ε is the eccentricity, and $\hat{\mathbf{L}}$ is a unit vector along the symmetry axis of the ellipsoid. We expect the elastic free energy per volume to have the form

$$\mathcal{F}_{el} = \frac{K}{R_c^2}\left(1 + \frac{1}{3}a_1 Q_{d_{\alpha\beta}}\Lambda_{\alpha\beta} + \ldots\right) \tag{13}$$

where $R_c^{-2} = \frac{1}{2}\langle R_1^{-2} + R_2^{-2}\rangle_d$ is mean squared curvature of the director field in the cavity, and a_i is a constant of the order of unity. The first term on the right hand side of Equation (13) is the elastic energy of the liquid crystal in a spherical inclusion; it is independent of the orientation of the droplet director. The second term is the contribution due to the non-spherical shape of the cavity. Higher order terms in the expansion correspond to higher moments of the cavity shape. Ignoring the orientation independent first term and letting $a_1/R_c^2 = 1/R_{\mathrm{eff}}^2$, the elastic free energy density can be written as

$$\mathcal{F}_{el} \simeq -\frac{1}{3}\frac{K}{R_{\mathrm{eff}}^2}\varepsilon^2 S S_d P_2(\hat{\mathbf{L}}\cdot\hat{\mathbf{N}}_d). \tag{14}$$

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Detailed calculations for certain director configurations confirm this result.[23] Thus, to lowest order, the effect of the cavity shape on the droplet order parameter is the same as that of an external field, and, for a prolate ellipsoid, the droplet direction $\mathbf{N}_d$ prefers to align parallel to the cavity axis $\hat{\mathbf{L}}$. In addition, we expect that other mechanisms, such as surface interactions, which may significantly contribute to the free energy and help determine the ground state configuration, will, for reasons of symmetry, give rise to terms of the form of Equation (14).

Orientation of droplet directors in the sample. The free energy per volume of a single droplet in an external field $\mathbf{H}$ is then given by

$$\mathcal{F} = -\frac{1}{3}\frac{K}{R_{\text{eff}}^2}\,\varepsilon^2 S S_d P_2(\hat{\mathbf{L}}\cdot\hat{\mathbf{N}}_d) - \frac{1}{3}\,\mu_0\Delta\chi H^2 S S_d P_2(\hat{\mathbf{H}}\cdot\hat{\mathbf{N}}_d). \tag{15}$$

If we further assume that S_d is a constant (independent of orientation and of the fields), then the free energy density of a single droplet can be written, to within an additive constant, in units of $K\varepsilon^2 S S_d/3R_{\text{eff}}^2$, as

$$\mathcal{F} = -P_2(\hat{\mathbf{L}}\cdot\hat{\mathbf{N}}_d) - h^2 P_2(\hat{\mathbf{H}}\cdot\hat{\mathbf{N}}_d) \tag{16}$$

where $h = (HR_{\text{eff}}/\varepsilon)\sqrt{\mu_0\Delta\chi/K}$ is the dimensionless applied field. The dimensionless magnetic field $h\hat{\mathbf{H}}$ and the cavity symmetry axis $\hat{\mathbf{L}}$ both act as fields, which, in general, compete to align the droplet director. Minimizing $\mathcal{F}$ with respect to $\hat{\mathbf{N}}_d$ in Equation (16) gives $\hat{\mathbf{N}}_d\cdot(\hat{\mathbf{H}}\times\hat{\mathbf{L}}) = 0$, and

$$P_2(\hat{\mathbf{H}}\cdot\hat{\mathbf{N}}_d) = \frac{1}{4} + \frac{3}{4}\frac{h^2 - 1 + 2(\hat{\mathbf{H}}\cdot\hat{\mathbf{L}})^2}{\sqrt{(h^2-1)^2 + 4h^2(\hat{\mathbf{H}}\cdot\hat{\mathbf{L}})^2}}. \tag{17}$$

If $h = 0$, then $P_2(\hat{\mathbf{H}}\cdot\hat{\mathbf{N}}_d) = P_2(\hat{\mathbf{H}}\cdot\hat{\mathbf{L}})$; that is, the droplet director $\hat{\mathbf{N}}_d$ is parallel to the cavity symmetry axis $\hat{\mathbf{L}}$. In the limit as $h \to \infty$, $P_2(\hat{\mathbf{H}}\cdot\hat{\mathbf{N}}_d) \to 1$; that is, the droplet director aligns parallel to the magnetic field. Equation (17) thus allows the calculation of the direction of the droplet director if h, $\hat{\mathbf{H}}$ and $\hat{\mathbf{L}}$ are known.

The sample order parameter may now be defined as $S_s = \langle \mathbf{P}_2(\hat{\mathbf{H}}\cdot\hat{\mathbf{N}}_d)\rangle_s$ where the average is over all droplets in the sample. S_s gives a measure of the alignment of the droplet directors in the sample; the degree of alignment of the liquid crystal molecules in the entire sample with the field is $\langle 3(\hat{\mathbf{H}}\cdot\hat{\mathbf{l}})^2 - 1\rangle_s = SS_dS_s$. Examples of the sample order parameter are shown in Figure 1(b). We assume that the orientational distribution of the symmetry axes $\hat{\mathbf{L}}$ of the droplets is uniform; then

$$S_s = \frac{1}{4\pi}\int P_2(\hat{\mathbf{H}}\cdot\hat{\mathbf{N}}_d)\,d\Omega_L = \frac{1}{4} + \frac{1}{16\pi}\int \frac{h^2 - 1 + 2(\hat{\mathbf{H}}\cdot\hat{\mathbf{L}})^2}{\sqrt{(h^2-1)^2 + 4h^2(\hat{\mathbf{H}}\cdot\hat{\mathbf{L}})^2}}\,d\Omega_L \tag{18}$$

where $d\Omega_L$ is an element of solid angle associated with the orientation of $\hat{\mathbf{L}}$. The integration gives

$$S_s = \frac{1}{4} + \frac{3(h^2+1)}{16h^2} + \frac{3(3h^2+1)(h^2-1)}{32h^3}\ln\left|\frac{h+1}{h-1}\right| \tag{19}$$

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The expression for the field dependence of the sample order parameter is our central result. The optical and dielectric response of the sample can be expressed in terms of this and as well as other order parameters.

The sample order parameter S_s is shown in Figure 2 as function of the dimensionless field h. Higher moments of the director distribution can be evaluated in the same way. In particular, the sample average of P_4 is

$$P_{4s} = \frac{7}{12} + \frac{5}{12} S_s - \frac{35}{32h^2} \cdot \left\{ \frac{2}{3} + \frac{(h^2 - 1)}{4h^2} - \frac{(h^2 + 1)^2(h^2 - 1)}{8h^3} \tan^{-1}\left(\frac{2h}{h^2 - 1}\right) \right\}. \tag{20}$$

Electric Field Induced Order

In the case of an applied electric field **E**, the free energy of each droplet cannot be considered independently because of droplet interactions via strong depolarizing fields. The electric field contribution to the free energy for the sample is

$$\mathcal{F}_E = -\frac{1}{2} \int_{\text{sample}} \mathbf{E} \cdot \mathbf{D} \, dV. \tag{21}$$

Exact evaluation of Equation (21) for a PDLC sample is prohibitively difficult. Instead, we use an effective medium approximation which has proven useful for describing the dielectric constant of these systems.[24] We imagine the real sample to be replaced by a homogeneous sample with the same dielectric characteristics.

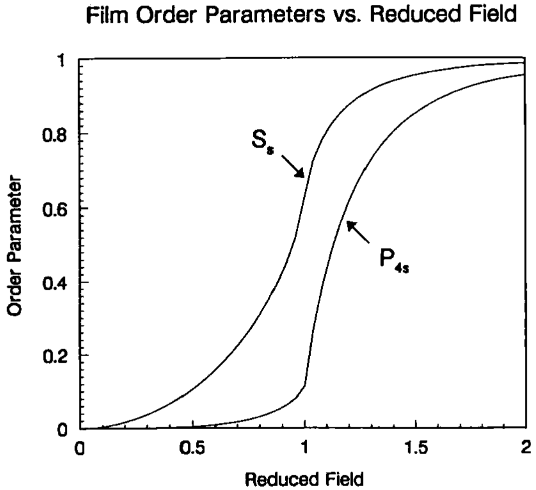
Film Order Parameters vs. Reduced Field

FIGURE 2 The sample order parameters S_s and P_{4s}, as a function of reduced field.

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A self-consistent calculation of the dielectric constant of the effective medium, ε_m, gives

$$\varepsilon_m = \varepsilon_p + 3v_{lc}\frac{\varepsilon_p(\varepsilon_{lc} - \varepsilon_p)}{\varepsilon_{lc} + 2\varepsilon_p - v_{lc}(\varepsilon_{lc} - \varepsilon_p)} \tag{22}$$

Here v_{lc} is the volume fraction of liquid crystal in the sample; ε_p is the dielectric constant of the polymer and ε_{lc} is the dielectric constant of the liquid crystal, averaged over the sample, in response to the applied field $\hat{\mathbf{E}}$. That is,

$$\varepsilon_{lc} = \langle \hat{\mathbf{E}} \cdot \underline{\boldsymbol{\varepsilon}}_{\mathbf{lc}} \cdot \hat{\mathbf{E}} \rangle_{\text{sample}} = \varepsilon_\perp + \frac{1}{3}\Delta\varepsilon + \frac{1}{N}\sum_i S_d \Delta\varepsilon P_2(\hat{\mathbf{E}} \cdot \hat{\mathbf{N}}_{di}) \tag{23}$$

The sum is over the N droplets in the sample. $\underline{\boldsymbol{\varepsilon}}_{\mathbf{lc}}$ is the dielectric tensor of the liquid crystal; $\varepsilon_\perp$ is the perpendicular dielectric constant of the liquid crystal and $\Delta\varepsilon$ is its dielectric anisotropy. If we again assume identical inclusions, with ellipsoidal shape and constant droplet order parameter, then

$$\varepsilon_{lc} = \varepsilon_\perp + \tfrac{1}{3}(1 + 2S_d S_s)\Delta\varepsilon \tag{24}$$

In terms of ε_m, the free energy density due to the field is

$$\mathcal{F}_E = -\tfrac{1}{2}\varepsilon_m E^2. \tag{25}$$

The evaluation of the sample order parameter is similar to the magnetic field case. The total free energy is minimized with respect to each droplet director $\hat{\mathbf{N}}_d$; then the sample order parameter is calculated by averaging over all cavity orientations. The result for S_s is identical in form to the magnetic field case (Equation 19), however, the dimensionless magnetic field h is replaced by a dimensionless electric field e:

$$e \equiv \frac{E g(S_s) R_{\text{eff}}}{\varepsilon}\sqrt{\frac{\Delta\varepsilon}{K}}, \tag{26}$$

where

$$g(S_s) = \frac{3\varepsilon_p}{\varepsilon_{lc} + 2\varepsilon_p - v_{lc}(\varepsilon_{lc} - \varepsilon_p)}. \tag{27}$$

The order parameter S_s is thus given implicity by Equation (19) in this case because of the dependence of e on S_s through ε_{lc} (cf. Equation 24). $g(S_s)$ is a dimensionless factor which describes the extent to which the applied field is shielded by depolarization charge built up at the droplet-polymer interface. If the dielectric constants are equal, $\varepsilon_{lc} = \varepsilon_p$, then $g = 1$. If g is only weakly dependent on S_s, then the dependence of the sample order parameter on the applied field is essentially the

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same for both electric and magnetic fields. In general, however, this is not the case. As an example, using typical values ($\varepsilon_p = 5\varepsilon_0$, $\varepsilon_\perp = 6\varepsilon_0$, $\Delta\varepsilon = 12\varepsilon_0$, and $v_{lc} = 0.4$) for a PMMA/E7 sample, we find $g(0) = 0.83$ and $g(1) = 0.65$. Thus there is more shielding of the applied field when the droplets are oriented by the field. This "negative feedback" results in a broader turn on region for the sample.

ORDER PARAMETER DYNAMICS

The dynamic response of system to time varying fields can be obtained by considering the torques acting on the liquid crystal in the droplets. Since the free energy can be expressed in terms of the orientation of the droplet director $\hat{\mathbf{N}}_d$, (Equation 18), the dynamical equation for $\hat{\mathbf{N}}_d$ is obtained by assuming that elastic and field induced torques are balanced by a viscous damping torque.[6] Then

$$\Gamma\dot{\Theta} = \frac{\partial}{\partial\Theta}(F_{\text{elastic}} + F_{\text{field}}) \tag{28}$$

where Θ is angle between the droplet director and the applied field, and Γ is a viscosity coefficient. Detailed analysis of the damping mechanism during reorientation has not been carried out; it is expected to originate from reorientation of the nematic director $\hat{\mathbf{n}}$ as the droplet configuration rotates. Elastic deformations in bipolar droplets are primarily bend and splay, and we assume that Γ is an average of the associated viscosity coefficients. Explicitly, the equation of motion is

$$\tau\dot{\Theta} = -\tfrac{1}{2}[\sin(2(\Theta - \gamma)) + f^2\sin(2\Theta)] \tag{29}$$

where γ is the angle between the applied field $\mathbf{E}$ and the symmetry axis $\hat{\mathbf{L}}$ of the cavity. Since the electric and magnetic field are formally equivalent, $f \equiv h$ or e designates the reduced field, and

$$\tau \equiv \frac{\Gamma R_{\text{eff}}^2}{K\varepsilon^2 S S_d} \tag{30}$$

is the characteristic time for the droplet.

The dynamic response can be determined by solving the equation of motion, Equation (29), with appropriate initial conditions, then the sample order parameter can be evaluated. Analytical results have been obtained for a number of special cases.

If the applied field varies slowly compared to τ, that is, if $\tau[\partial \ln f(t)/\partial t] << 1$, the viscous torque can be neglected. The response is then quasistatic, and the order parameters are the same as for the static case but with constant field replaced by the time dependent $f(t)$.

The situation where a constant field f_0 is suddenly applied or removed is of

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considerable practical importance. It is convenient to write Equation (29) in this case as

$$\dot{\Theta} = \frac{1}{2\tau}\cdot\frac{\sin(2\gamma)}{\sin(2\alpha)}\sin 2(\Theta - \alpha) \tag{31}$$

where

$$\sin(2\alpha) \equiv \frac{\sin(2\gamma)}{\sqrt{\sin^2(2\gamma) + (f_0^2 + \cos(2\gamma))^2}} \tag{32}$$

Integration of Equation (31), with initial condition $\Theta = \Theta_0$ at $t = 0$, gives

$$\tan(\Theta - \alpha) = \tan(\Theta_0 - \alpha)e^{-\sin 2\gamma/\sin 2\alpha \cdot t/\tau}. \tag{33}$$

The time dependence of the order parameter, $S_s(t)$, can be obtained by noting that

$$\cos^2\Theta = \frac{(1 - \omega\tan\alpha)^2}{(1 + \omega^2)(1 + \tan^2\alpha)}, \tag{34}$$

where

$$\omega \equiv \tan(\Theta - \alpha). \tag{35}$$

Then, from Equation (18),

$$S_s(t) = -\frac{1}{2} + \frac{3}{8\pi}\int_{\Omega_L}\frac{(1 - \omega\tan\alpha)^2}{(1 + \omega^2)(1 + \tan^2\alpha)}\,d\Omega_L \tag{36}$$

and similarly

$$\langle P_4(t)\rangle_s = -\frac{7}{8} - \frac{5}{2}S_s(t) + \frac{35}{32\pi}\int_{\Omega_L}\left[\frac{(1 - \omega\tan\alpha)^2}{(1 + \omega^2)(1 + \tan^2\alpha)}\right]^2 d\Omega_L. \tag{37}$$

For an arbitrary applied field f_0 and initial distribution of droplet directors, Equations (36) and (37) must be evaluated numerically. A special case of considerable importance is the case where initially $S_s = 1$, due to a large applied field f_0 which is removed at $t = 0$. The optical transmittance of a PDLC sample in this case will decrease with time as the directors relax to the zero field equilibrium states. This decay in transmittance is an important material property for light shuttering applications. Setting $f_0 = 0$ and $\Theta_0 = 0$ gives $\alpha = \gamma$, and

$$\omega = -\tan(\gamma)e^{-t/\tau}. \tag{38}$$

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Equations (36) and (37) can be integrated to give

$$S_s(t) = 1 - \frac{1}{(1+R)^2}\left[1 + \frac{3R^2}{2} - \frac{3R}{2\sqrt{1-R^2}}\tan^{-1}\left(\frac{\sqrt{1-R^2}}{R}\right)\right], \quad (39)$$

and

$$\langle P_4(t)\rangle_s = -\frac{7}{8} - \frac{5}{2}S_s(t) + \frac{1}{(1+R)^4}\left[\frac{7}{8} + \frac{35R}{6} + \frac{1421R^2}{48} + \frac{35}{3}R^3 \right.$$
$$\left. + \frac{7R^4}{3} + \frac{35(8R+1)R}{16\sqrt{1-R^2}}\, a \tan\left(\frac{\sqrt{1-R^2}}{R}\right)\right], \quad (40)$$

where

$$R = e^{-t/\tau}. \quad (41)$$

This decrease of order with time is shown in Figure 3. Unlike the response of a single droplet, the decay is not simply exponential, except at very late times ($t >> \tau$).

For a periodic applied field $f(t) = f_0 \sin(\omega t)$, the droplet directors oscillate about some average direction Θ_0, and we write

$$\Theta(t) \equiv \Theta_0 + \Delta(t). \quad (42)$$

The equation of motion, Equation (29) gives, to lowest order in Δ,

$$\dot{\Delta} = -\frac{1}{2\tau}[\sin(2(\Theta_0 - \gamma)) + f^2 \sin(2\Theta_0)]. \quad (43)$$

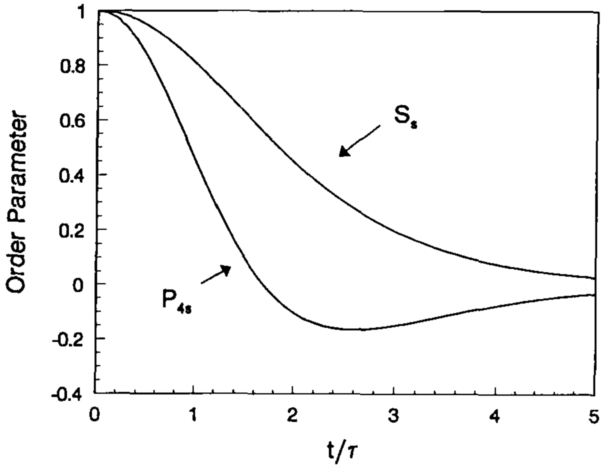

FIGURE 3 Decay of the film order parameters S_s and P_{4s} with time after removal of a strong applied field.

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Taking the time average of both sides gives

$$\sin(2(\Theta_0 - \gamma)) + \langle f^2 \rangle \sin(2\Theta_0) = 0 \tag{44}$$

and the average angle Θ_0 of the droplet director is the same as the equilibrium response to the root mean square value of the applied field. Equations (43) and (44) give

$$\Delta(t) = \frac{-1}{4\omega\tau} \sin(2(\Theta_0 - \gamma))\sin(2\omega t) \tag{45}$$

As expected, the response is at 2ω, and the oscillations are small when $\omega\tau >> 1$. At high frequencies therefore the order parameters are given by their equilibrium expressions with the static field replaced by its root mean square value.

APPLICATION OF THE MODEL: LIGHT TRANSMISSION IN PDLC SAMPLES

The optical and dielectric properties of PDLC materials are related to the sample order parameters. An expression for the dielectric constant has already been given in Equations (22) and (24). Next, we consider the optical transmittance of PDLC materials in some detail.

The intensity of light traversing a PDLC film is attenuated as light is scattered out of the beam by the liquid crystal droplets. Ignoring multiple scattering effects, the intensity $I(z)$ of the beam after transversing a distance z in the sample is

$$I(z) = I_0 e^{-\rho_d \sigma_s z}. \tag{46}$$

where ρ_d is the number density of droplets and σ_f is the average droplet scattering cross-section, averaged over the sample.

We are interested in the transmittance of PDLC films as a function of a field applied normal to the film. If a plane-polarized beam of monochromatic light of wavelength λ is incident normal to the film plane, its electric field is

$$\mathbf{E} = \mathbf{E}_0 e^{i(\mathbf{k}\cdot\mathbf{r} - \omega t)} \tag{47}$$

Each droplet director makes an angle Θ with respect to the applied field. The total cross-section for a given droplet will depend on the relative orientations of the three vectors $\mathbf{k}$, $\mathbf{E}_0$, and $\hat{\mathbf{N}}_d$. Since the projection of the droplet directors onto the plane normal to $\mathbf{k}$ is random, the droplet cross-section can only be a function of the angle $\hat{\mathbf{N}}_d \cdot \mathbf{k} = \cos\Theta$. Expanding in Legendre polynomials,

$$\langle \sigma_d \rangle_E = \sum_{l=0}^{\infty} a_{2l} P_{2l}(\cos\Theta). \tag{48}$$

$\langle . . . \rangle_E$ indicates that the dependence on the orientation of the electric vector has

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been averaged out. Only even order terms are admissible since there is no physical difference between $\hat{\mathbf{N}}_d$ and $-\hat{\mathbf{N}}_d$. The detailed information about the droplet cross-section—its dependence on shape, size, wavelength and director configuration—is contained in the expansion coefficients. Averaging over all droplets,

$$\sigma_s = \sum_{l=0}^{\infty} a_{2l} \langle P_{2l}(\cos \Theta) \rangle_s. \tag{49}$$

The quantities in brackets are just the sample order parameters. Although all even terms in the expansion are required to specify σ_s exactly, we show below that the first few terms give an adequate description of the transmitted light intensity in some typical situations.

Zumer[25] and Zumer and Doane[17] have looked in detail at the scattering of light from a single spherical liquid crystal droplet. The droplet is presumed to be embedded in an isotropic matrix with a comparable refractive index. We now combine their results with our order parameter formalism.

Rayleigh-Gans Scattering

We first consider small droplets, where the droplet diameter d is much smaller than the wavelength of light λ. Here the Rayleigh-Gans approximation[26] (RGA) is valid. A good description of the scattering can be provided in terms of the droplet order parameter in the regime where $kd << 1$ and $k = 2\pi/\lambda$.

In general, the differential scattering cross-section, $d\sigma/d\Omega$, can be expressed in terms of the scattering amplitude $\mathbf{f}(\mathbf{k}, \mathbf{k}')$ and the incident field $\mathbf{E}_0$ by

$$\frac{d\sigma}{d\Omega} = |\mathbf{f}(\mathbf{k}, \mathbf{k}')|^2/|\mathbf{E}_0|^2, \tag{50}$$

where $\mathbf{k}$ and $\mathbf{k}'$ are the incident and scattered wavevectors, respectively. In the RGA, the scattering amplitude is given by

$$\mathbf{f}(\mathbf{k}, \mathbf{k}') = \frac{1}{4\pi} V_d k^2 (\mathbf{P} - \hat{\mathbf{i}}'[\hat{\mathbf{i}}' \cdot \mathbf{P}]). \tag{51}$$

where V_d is the volume of the droplet, $\hat{\mathbf{i}}' \equiv \mathbf{k}'/k$, and

$$\mathbf{P} = \mathbf{E}_0 \cdot \langle (\underline{\boldsymbol{\varepsilon}}_r - \underline{\mathbf{1}}) e^{-i\mathbf{k}_s \cdot \mathbf{r}} \rangle_d \tag{52}$$

where $\langle \ldots \rangle_d$ denotes a volume average over the droplet, $\mathbf{k}_s = \mathbf{k}' - \mathbf{k}$ is the scattering vector, and the relative dielectric tensor is defined in its local principal axis frame by

$$\underline{\boldsymbol{\varepsilon}}_r = \frac{1}{\varepsilon_p} \begin{bmatrix} \varepsilon_\perp & 0 & 0 \\ 0 & \varepsilon_\perp & 0 \\ 0 & 0 & \varepsilon_\parallel \end{bmatrix}. \tag{53}$$

ε_p is the dielectric constant of the (polymer) matrix, and $\varepsilon_\parallel$ is the dielectric constant for fields parallel to the local director $\hat{\mathbf{n}}(\mathbf{r})$.

If $kd << 1$, $e^{i\mathbf{k}_s \cdot \mathbf{r}} \approx 1$, and Equations (52) gives

$$\mathbf{P} \approx \mathbf{E}_0 \cdot \langle \underline{\boldsymbol{\varepsilon}}_r - \underline{\mathbf{1}} \rangle_d. \tag{54}$$

In this approximation, the droplet is a radiating dipole whose polarizability is the volume averaged polarizability of the droplet. Following the notation of Zumer and Doane,[17] we separate the dielectric tensor into an isotropic and an anisotropic part:

$$\underline{\boldsymbol{\varepsilon}}_r - \underline{\mathbf{1}} = \zeta \underline{\mathbf{1}} + \frac{\eta}{s} \underline{\mathbf{Q}}, \tag{55}$$

where

$$\zeta = \tfrac{1}{3} Tr(\underline{\boldsymbol{\varepsilon}}_r) - 1, \tag{56}$$

and

$$\eta = \frac{\varepsilon_\parallel - \varepsilon_\perp}{3\varepsilon_p}. \tag{57}$$

$\underline{\mathbf{Q}}$ is the local order parameter tensor. It follows at once that

$$\langle \underline{\boldsymbol{\varepsilon}}_r - \underline{\mathbf{1}} \rangle_d = \zeta \underline{\mathbf{1}} + 2\eta \underline{\mathbf{Q}}_d, \tag{58}$$

where $\underline{\mathbf{Q}}_d$ is the droplet order parameter tensor. We note that if we make the formal replacement $\eta \rightarrow S_d\eta$, subsequent development is the same as that of Zumer and Doane[17] for a uniform director configuration ($S_d = 1$) in the limit of $kd <<$ 1. In particular, the total scattering cross-section is found to be

$$\sigma_d^{rga} = \sigma_0 \frac{(kd)^4}{108} \cdot [(\zeta - \eta S_d)^2 + 3\eta S_d(2\zeta + \eta S_d)\cos^2\alpha_0 \sin^2\Theta]. \tag{59}$$

Here $\sigma_0 = \pi d^2/4$ is an effective geometrical cross-section for the droplet, and α_0 is the angle between the incident electric field and the plane defined by $\hat{\mathbf{N}}_d$ and $\mathbf{k}$. The scattering cross-section of the droplet shows the $1/\lambda^4$ dependence characteristic of Rayleigh scattering. We have carried out detailed comparisons of Equation (59) with the numerical results for a variety of director configurations[17]; the agreement is within 1% for $kd < 0.3$ and within 10% for $kd < 1.0$. Averaging over all orientations of the incident electric field as before, $\langle \cos^2\alpha_0 \rangle = \frac{1}{2}$, and

$$\langle \sigma_d^{rga} \rangle_E = \sigma_0 \frac{(kd)^4}{108} \cdot [(\zeta - \eta S_d)^2 + \frac{3}{2} \eta S_s(2\zeta + \eta S_d)\sin^2\Theta]. \tag{60}$$

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Finally, to obtain the average sample cross-section, we average overall droplet director orientations in the film. This gives

$$\sigma_s^{rga} = \sigma_0 \frac{(kd)^4}{108} \cdot [((\zeta - \eta S_d)^2 + \eta S_d (2\zeta + \eta S_d) \cdot (1 - S_s)] \tag{61}$$

Equation (61) gives the average scattering cross section for droplets in the sample in terms of the droplet and sample order parameters; it is expected to hold when the RGA is valid. When $S_d = 1$, the second term in the brackets vanishes. In this case, the cross-section vanishes when

$$\zeta = \eta S_d. \tag{62}$$

This is the index matching criterion for samples with small droplets.

Anomalous Diffraction Regime

For droplets larger than a few tenths of a micron, the anomalous diffraction approach (ADA) gives a better description of the scattering than the Rayleigh-Gans; however, analytical results are then restricted to the case of a uniform director configuration. We expect that, for the purposes of calculating the scattering cross-section for bipolar droplets, the uniform director configuration is an acceptable approximation. We also restrict the analysis to the case where $kd < 10$; in this regime, it is possible to obtain detailed analytical results. For optical wavelengths, this restricts the droplet size to $d < 1$ micron. The total droplet cross-section for a sphere with a uniform configuration in the ADA is,[25] for small kd,

$$\sigma_d^{ada} = \frac{1}{2} \sigma_0 \cdot (kd)^2 \cdot \left[\cos^2\alpha_0 \cdot \left[\frac{n_e(\Theta)}{n_p} - 1 \right]^2 + \sin^2\alpha_0 \cdot \left[\frac{n_o}{n_p} - 1 \right]^2 \right]. \tag{63}$$

Here n_o and n_e are the ordinary and extraordinary indices of the liquid crystal and n_p is the index of the polymer matrix. Θ, σ_0, and α_0 have been defined earlier; and

$$n_e(\Theta) = \left[\frac{\cos^2\Theta}{n_o^2} + \frac{\sin^2\Theta}{n_e^2} \right]^{-1/2}. \tag{64}$$

Expanding $n_e(\Theta)$ in terms of $\Delta n \equiv n_o$ gives, to lowest order,

$$n_e(\Theta) \approx n_o + \Delta n \cdot \sin^2\Theta. \tag{65}$$

Averaging σ_d^{ada} over field and droplet director orientations as before obtains the

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average scattering cross section of the droplets in the sample in terms of the sample order parameter S_s and $\langle P_4 \rangle_s$,

$$\sigma_s^{ada} = \frac{1}{2}\sigma_0 \cdot (kd)^2 \cdot \left(\frac{\Delta n}{n_p}\right)^2 \cdot \left[\left(\frac{\delta}{\Delta n}\right)^2 - \frac{2}{3}\cdot\frac{\delta}{\Delta n}\cdot(1 + S_s) + \frac{4}{105}\cdot(7 - 10S_s + 3\cdot\langle P_4\rangle_s)\right]. \quad (66)$$

Here $\delta \equiv n_p - n_0$ is a measure of the mismatch between the index of the polymer and the ordinary index of the liquid crystal. When $\delta = 0$, σ_s^{ada} is a monotonically decreasing function of S_s and vanishes when the droplets are parallel to the applied field. If $\delta < 0$, σ_s^{ada} is again monotonically increasing but does not vanish when $S_s = 0$. If $\delta > 0$, then σ_s^{ada} vanishes at $S_s < 1$ and is nonzero when the droplets are completely aligned. The three situations are shown in Figure 4 for typical sample parameters. This behavior has been observed in certain PDLC films.[27] In contrast, we note that σ_s^{rga} increases monotonically in all cases; the transmission does not show similar peaking for non-index-matched films containing small droplets.

Equation (66) indicates a quadratic wavenumber dependence for the dispersion in the ADA. In fact, deviations from this quadratic dependence in the detailed numerical results[25] at higher values of kd can be used to establish an upper limit for the validity of Equation (66). This gives $kd \simeq 10$ as a cutoff.

Next, we calculate the transmittance in the AD regime of a PDLC film composed of E7 and the thermoplastic polymer polymethylmethacrylate (PMMA) as a function of applied voltage. Experimental results together with predictions of the model are shown in Figure 5. Index matching was assumed in the calculation, and the

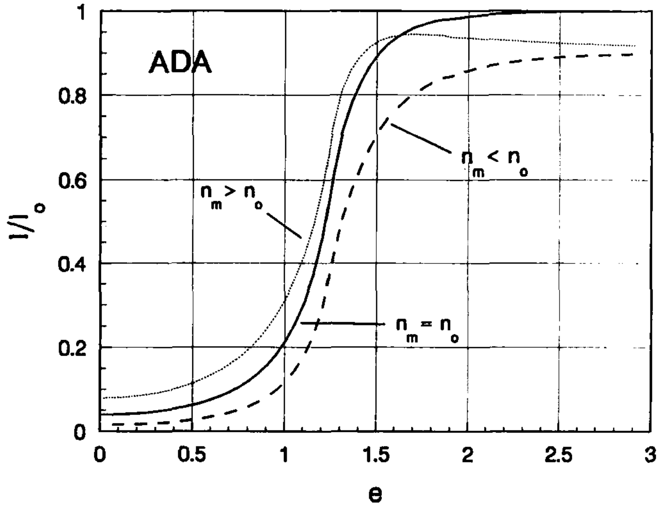

FIGURE 4 Calculated sample transmittance versus field for different mismatch between the polymer refractive index n_p, and the ordinary index of the liquid crystal, n_o.

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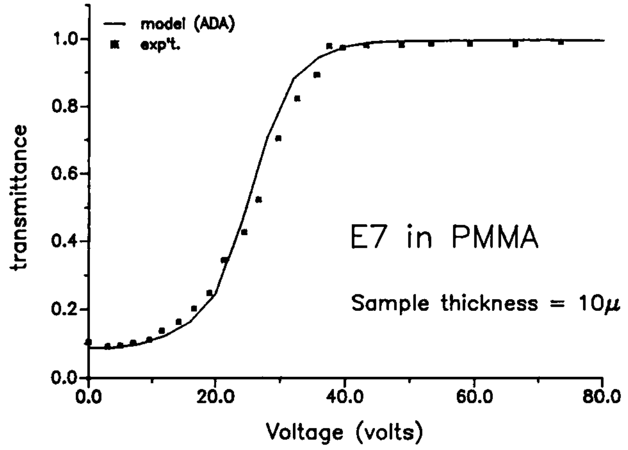

FIGURE 5 Optical transmittance of a 10-μm thick PMMA/E7 PDLC film as function of applied voltage in the anomalous diffraction regime.

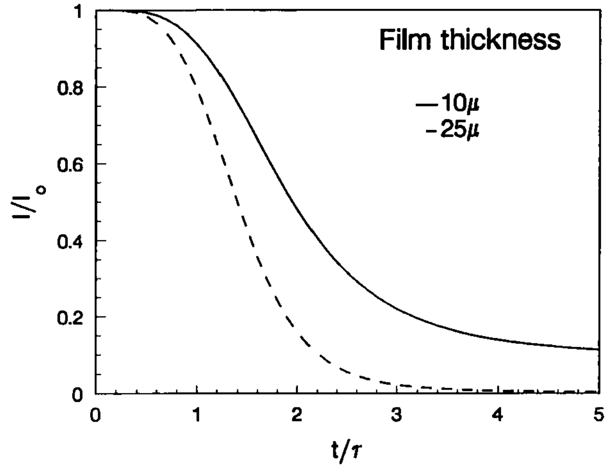

FIGURE 6 Calculated decay of sample transmittance as function of time with time, for two different film thicknesses using the same material characteristics as in Figure 5.

zero-field value of the exponent $\rho_d \sigma_s z$ in Equation (46) was chosen to match the zero-field transmittance from the data. The shape of the curve was then completely determined. The model curve was scaled to achieve a match of the two curves at 50% transmittance. This scaling was necessary because the parameters of the reduced field, particularly the droplet eccentricity, are not sufficiently well known.

The time dependence of the transmittance can be obtained by using the time dependent expressions for the sample order parameter in the expressions for the scattering cross section. The calculated decay of the transmittance for the PMMA/E7 sample is shown in Figure 6 for two different film thicknesses. It is interesting to note that the turn-off time is nearly 50% greater for the thinner film.

CONCLUSIONS

Polymer dispersed liquid crystals are complex materials with aspects which are incompletely understood. Rather than attempting a detailed description, we have proposed a simple approximate model which provides a framework for the interpretation and prediction of the optical behavior. The model relates the optical and dielectric response of these materials to external fields via their effect on a hierarchy of order parameters. This approach may be useful in describing other complex materials.

Results for the average droplet scattering cross-sections in terms of the order parameters give a good description of the transmittance of a function of applied field. σ_s^{rga} and σ_s^{ada}, given by Equations (61) and (66), are sufficient to describe the scattering from nematic droplets over the range $0 < kd < 20$. The crossover occurs in the region near $kd \approx 2$. Terms up to P_4 in the expansion for the cross-section are sufficient to model the response in this range. In other cases, we anticipate that it may be necessary to include terms of higher order, or to make use of the distribution function directly.

The assumption that the droplet directors are randomly distributed is realistic for PDLC materials formed by phase separation techniques. However, films formed by encapsulation methods (NCAP) typically have their major axes distributed near the film phane. In this case, it is possible to formulate a response model based on the distribution of critical switching fields for droplets of different sizes.[9]

The model has been used to interpret dielectric studies and the wavelength dependence of light transmission in PDLC films.[28] The model can also be easily extended to include non-uniform droplet size distributions[11]; multiple scattering has also been considered.[29] Studies to compare predictions of the model with the measured response in other PDLC materials are under way. These results will appear elsewhere.

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