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Theory of Dielectric and Optical Properties of PDLC films

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We present results of the light scattering properties of PDLC films in the Rayleigh-Gans long wavelength régime. The calculations take into account dependent scattering effects using an effective medium theory. The Percus-Yevick hard-sphere approximation is used to introduce droplet correlation effects. We discuss the light scattering properties of radial and bipolar partially ordered droplets, making detailed comparisons with the literature where appropriate.

Keywords: PDLC films; effective medium theory; bipolar droplets; Percus-Yevick; light scattering; inhomogeneous media.

INTRODUCTION

We have made a theoretical study of the light scattering properties of Polymer Dispersed Liquid Crystal (PDLC) films. These devices have a number of applications including direct view and projection display technology, switchable windows, electro-optic shutter devices, large-scale flexible displays, and high resolution active matrix addressing systems.[1, 2, 3]

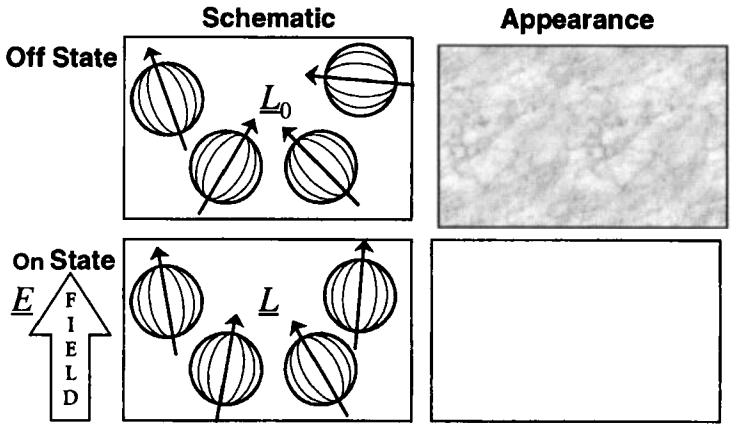


FIGURE 1 Schematic representation of a PDLC film.

A PDLC film, shown schematically in figure 1, consists of a random ensemble of micrometre-sized liquid crystal droplets dispersed in an isotropic polymer matrix. In the absence of any external electric field, the film has a milky white translucent appearance. However, when a field is applied, the film becomes clear. In contrast to polarization devices, there is little loss of light in the transparent state, making them highly desirable. The contrast between the off and on states relies on an optical mismatch between the liquid crystal droplets and the polymer matrix, which disappears when the field is applied.

In a recent series of papers[4,5,6], we construct a detailed self-consistent effective medium theory of the dielectric properties of PDLC films, and considered their light scattering properties in the Rayleigh-Gans régime. In this paper, we shall briefly review this approach, compare it with other methods employed in the literature[7,8,9], and discuss the technologically important case of partially ordered bipolar systems. Here we emphasise the quantitative consequences of important assumptions in the model; for the mathematical detail we refer readers elsewhere [4,5,6].

THEORY

A theory of light scattering from partially ordered droplet systems requires several components[4,5,6]:

- A theory of the single scattering by individual liquid crystal droplets.
- A structure factor to describe the positional correlations of droplets.
- An understanding of the reorientation of droplets in the presence of an electric field, and the effect of this has on the film's optical characteristics.

In addition the problem is further complicated by:

- Shape and size polydispersity.
- Multiple scattering effects.

Types of droplet

Figure 2 shows the two types of droplet which we consider[4].

FIGURE 2 (a) Radial droplets (b) Bipolar droplets.

Radial droplets are indistinguishable; the director is strongly anchored in a radial direction at the droplet boundary. Bipolar droplets contain a pair of surface boojum defects at their poles. These define a local orientation axis, $\underline{L}_0$, for a given droplet. We refer to a system of such droplets as *partially ordered.* We may impose order on the droplet directors by the application of an electric field. We will show that a partially ordered system of bipolar droplets has the characteristics required to make a technologically viable PDLC film.

Optical Properties of PDLC films

We define the dielectric constants of the material at the relevant optical frequencies as

$$\begin{aligned} \epsilon_{\parallel} &= \epsilon_{is} + \epsilon_a \\ \epsilon_{\perp} &= \epsilon_{is} - \tfrac{1}{2}\epsilon_a \end{aligned} \qquad \begin{cases} \epsilon_{is} = \tfrac{1}{3}\left(\epsilon_{\parallel} + 2\,\epsilon_{\perp}\right) \\ \epsilon_a = \tfrac{2}{3}\left(\epsilon_{\parallel} - \epsilon_{\perp}\right) \end{cases} . \tag{1}$$

Our definition ϵ_a is slightly unconventional. We use the following numerical values for the optical dielectric constants in our calculations:

$$\begin{aligned} \epsilon_{\parallel} &= n_e^2 = 1.7^2 \\ \epsilon_{\perp} &= n_o^2 = 1.52^2 \\ \epsilon_m &= n_m^2 = 1.55^2 \end{aligned} , \tag{2}$$

where n_e and n_o are the ordinary and extraordinary refractive indices of a typical PDLC droplet[7] and n_m is the refractive index of the polymer matrix.

Droplet Re-orientation in the Presence of a Field

The details of this process appear elsewhere[5]. What follows is a qualitative review. For the bipolar droplets, there is a local orientation axis, $\underline{L}_0$, define by the surface defects (see figure 2). In the presence of an electric field, the droplet axes realign, as shown schematically in figure 1. We assume that the applied field only affects the inclination of the bipolar axis to the field, not its azimuthal angle. The azimuthal angles of the droplets are randomly distributed in the interval [0, 2π]. The realignment of a droplet depends on (i) its initial alignment with the field (ii) the droplet concentration. As the droplet concentration increases, a cooperative effect makes all droplets realign at a lower critical field, $\tilde{U}_c$. The applied field is measured in terms of a non-dimensionalised voltage, $\tilde{U}$, where, $\tilde{U}$ = 1 corresponds to about 2.5 Volt. This phenomenological theory contains a droplet pinning parameter, W, which measures the balance between the elastic energy from the initial alignment of a droplet and the applied field. We will return to this later.

We may describe the re-orientation of a system of droplets, initially aligned randomly with the field, in terms of an order parameter for the bipolar axes, Q, (c.f. S_s used by Kelly and Palffy-Muhoray[9]). Q makes a sharp transition from 0 to 1 at the transition voltage $\tilde{U}_c(\eta)$. We may determine the light-scattering properties of the droplet in terms of Q and a higher order spherical harmonic Q_4, which are derived numerically from averages over the distribution of droplet alignments to the field at a given voltage, $\tilde{U}$.

Light Scattering in Inhomogeneous Media

For a more comprehensive study of scattering theory see, for example, Ishimaru[10], Newton[11], Bohren and Huffman[12], and van de Hulst[13]. This section contains a brief review of the theory that we require.

A PDLC film is an inhomogeneous medium consisting of droplets embedded in a polymer matrix. The droplets we consider are spherical, though in principal we could consider more general ellipsoidal droplets. In all calculations, we have considered unpolarised light (which is involves averaging over light polarisations) and the light impacts the PDLC film at normal incidence. We will consider the important case of non-normal incidence elsewhere. Important notation, formulae and observable quantities are defined in Table I.

By analogy with the scattering of, for example, neutrons, we consider the scattering to consist of two parts:

1. *Coherent scattering*, I_C: is scattering by an average droplet, and is reduced at high droplet concentrations by destructive interference.
2. *Incoherent scattering*, I_I: is scattering caused by uncorrelated orientational fluctuations. It is additive and not reduced at high droplet concentrations.

TABLE I Important light scattering formulae, and notation.

Scatterer size	a
Droplet volume fraction	η
Number density of droplets	$\rho = \frac{3}{4\pi a^3}\eta$
Momentum Transfer	$q = 2k\sin\left(\frac{\theta_s}{2}\right)$
Droplet structure factor	$S(q)$
Coherent and Incoherent Scattering	$I_C(\underline{k},\underline{k}')$ and $I_I(\underline{k},\underline{k}')$
Effective differential cross-section	$\frac{\partial\sigma}{\partial\Omega} = S(q)\,I_C(\underline{k},\underline{k}') + I_I(\underline{k},\underline{k}')$
Scattering cross section	$\sigma_s = \int \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\,\mathrm{d}\Omega$
Inverse scattering mean free path, attenuation coefficient	$l_s^{-1} = \mu = \rho\sigma_s$
Film thickness	D
Beam intensity	$I = I_0\exp(-\mu D)$
Single particle transport cross section	$\sigma_{tr} = \int \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\left(1-\cos(\theta_s)\right)\mathrm{d}\Omega$
Inverse transport mean free path	$l_{tr}^{-1} = \rho\sigma_{tr}$

Percus-Yevick hard spheres approximation

There are two common approaches to derive the structure factor required in Table I. This modifies the coherent scattering at angle θ_s by a factor $S_0(q)$. The structure factor can be related to the positional distribution of the scatterers:

$$g(\underline{r}) = 1 + \rho\int d\underline{r}\,\left(S(\underline{q}) - 1\right)\exp\left\{-i\underline{q}.\underline{r}\right\}, \qquad (3)$$

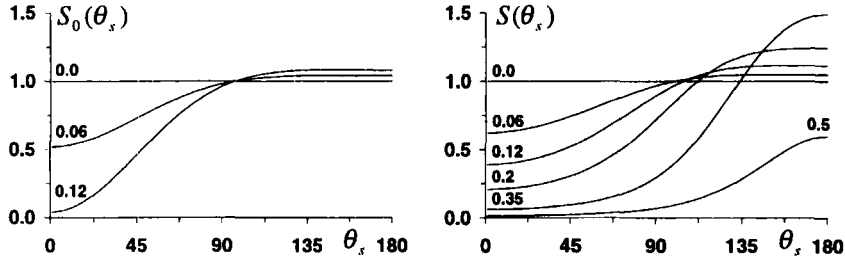
where $g(\underline{r})$ is the factor which, as a result of positional correlations, modifies the local scatterer density at a point $\underline{r}$, given that a scatterer is placed at the origin.

Žumer *et al* [8] adopts the hole approximation to describe the correlations between the droplet positions, which prescribes only that droplets of diameter d do not overlap. This gives the structure factor $S_0(q)$:

$$S_0(q) = 1 + 24\eta \frac{2qa\cos(2qa) - \sin(2qa)}{(2qa)^3}; \qquad (4)$$

which becomes unphysical (negative) for droplet concentrations above η = 1/8.

We have adopted a more sophisticated approximation based on the Percus-Yevick theory of liquids[6, 14, 15, 16, 17] in which the droplets are treated as hard spheres. This allows us to study the effects of droplet concentration on the light scattering properties of a PDLC film up to $\eta \sim 0.55$, which is the point at which the system is likely to become crystalline. It is unlikely that a PDLC film with such high droplet concentrations could be fabricated. In figure 3, we compare the two structure factors for ka = 1.5.



(a) Hole approximation, $S_0(q)$ (b) Percus-Yevick approximation.

FIGURE 3 Structure factors for ka = 1.5.

Both structure factors decrease the forward scattering. The hole approximation has a more dramatic effect at lower values of η than the Percus-Yevick structure factor.

Effective Medium Theory

Our light scattering formulae are only strictly valid for small η: as η increases, the effective scattering amplitude will be affected by the proximity of other scatterers. This is sometimes known as dependent scattering.

We replace the dielectric constant of the polymer matrix, ϵ_m, by an effective dielectric constant, $\underline{\bar{\epsilon}}$, which takes into account both the underlying polymer matrix, and also the dielectric properties of the other droplets in the film. In general, $\underline{\bar{\epsilon}}$ is a tensor.

The effective medium theory which we have adopted[4,5,6] is the coherent potential approximation[18], which is one of the Maxwell-Garnett family of effective medium theories[19, 20]. We suppose that the liquid crystal dielectric constant within a droplet can be replaced by its mean value, which gives[5]:

$$\underline{\bar{\epsilon}} = \epsilon_m \underline{\underline{I}} + \eta \sum_i p_i \left(\underline{\bar{\epsilon}}_i - \epsilon_m \underline{\underline{I}}\right)\left(\tfrac{2}{3}\underline{\underline{I}} + \tfrac{1}{3}\epsilon_m^{-1}\underline{\bar{\epsilon}}_i\right)^{-1}, \tag{5}$$

where the sum is taken over the probability, p_i, of encountering droplets with tensor dielectric constant $\underline{\bar{\epsilon}}_i$. $\underline{\underline{I}}$ is the unit tensor. To calculate the realignment of droplets in a film, we use the full tensor $\underline{\bar{\epsilon}}$, but to study the light scattering properties of the film, we must adopt a scalarised form. In the case of isotropic scatters, this is straightforward, since $\underline{\bar{\epsilon}}_i = \epsilon_{lc} \underline{\underline{I}}$:

$$\bar{\epsilon} = \frac{\epsilon_m\left(\epsilon_{lc} + 2\epsilon_m + 2\eta\left(\epsilon_{lc} - \epsilon_m\right)\right)}{\left(\epsilon_{lc} + 2\epsilon_m - \eta\left(\epsilon_{lc} - \epsilon_m\right)\right)}. \tag{6}$$

In the case of the partially ordered system, we interpolate to obtain an effective scalar value from $\underline{\bar{\epsilon}}$, which we shall justify *ex post facto*.

$$\bar{\epsilon} = (1-\eta)\epsilon_m + \eta\,\epsilon_{lc}. \tag{7}$$

RESULTS

In this section we compare our results with those of Žumer *et al* [8] for the case of radial droplets, demonstrating quantitatively the affects of each part of our theory. We have also made similar careful comparisons in the case of droplets in a strong field which yield results which may be understood analogously. We will then use our unified theory to derive further results in the technologically important case of partially ordered bipolar droplets. For completeness, we have included formulae derived elsewhere[6].

Radial Droplets

In the case of a system of radial droplets, show schematically in figure 2, all of the droplets are identical, and hence there is no incoherent scattering. Our formulae for the scattering are[6]:

$$I_C(\theta_s) = \frac{a^6 k^4}{18}\left(|\hat{\epsilon}_{xx}|^2 + |\hat{\epsilon}_{yy}|^2 + |\hat{\epsilon}_{zy}|^2 - |\hat{\epsilon}_{zy}\cos(\theta_s) + \hat{\epsilon}_{yy}\sin(\theta_s)|^2 \right)$$

$$\begin{aligned}
\hat{\epsilon}_{xx} &= \hat{\epsilon}_I - \tfrac{1}{2}\hat{\epsilon}_A \\
\hat{\epsilon}_{yy} &= \hat{\epsilon}_I + \tfrac{1}{2}\hat{\epsilon}_A\left(3\cos^2\left(\tfrac{\theta_s}{2}\right) - 1\right) \\
\hat{\epsilon}_{zy} &= -\tfrac{3}{2}\hat{\epsilon}_A\cos\left(\tfrac{\theta_s}{2}\right)\sin\left(\tfrac{\theta_s}{2}\right) \\
\hat{\epsilon}_I &= \frac{3}{(qa)^3}\left(\frac{\epsilon_{is}}{\bar{\epsilon}} - 1\right)\left(qa\cos(qa) - \sin(qa)\right) \\
\hat{\epsilon}_A &= \frac{3}{(qa)^3}\left(\frac{\epsilon_u}{\bar{\epsilon}}\right)\left(qa\cos(qa) - 4\sin(qa) + 3\mathrm{Si}(qa)\right)
\end{aligned} \tag{8}$$

Hole Approximation

Using the hole approximation structure factor, (4), and the parameters $\left(\frac{\epsilon_{is}}{\bar{\epsilon}} - 1\right) = 0.04$ and $\left(\frac{\epsilon_u}{\bar{\epsilon}}\right) = 0.16$, we obtain, in figure 4(a), the same results as Žumer *et al* [8] for the differential scattering cross-section for various values of the packing fraction, η. Rounding the values for these parameters

introduces a change of ~10 % in the calculations, which would mask the change as the effective medium approximation introduces. In figure 4(b) we repeat the calculations using the "exact" values of (2) in (1), and using $\overline{\epsilon} = \epsilon_m$. Without the rounding, we see that the differential cross-section is slightly increased.

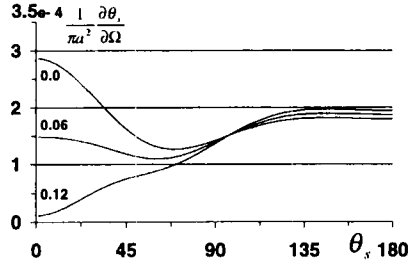
Effective medium approximation

As the droplet concentration increases, the dielectric properties of the effective medium, $\overline{\epsilon}$, become less like the polymer matrix, ϵ_m and more like the droplets, ϵ_{lc}. For radial droplets, we use $\epsilon_{lc} = \epsilon_{is}$ and $\overline{\epsilon}$ from (6). Figure 4(c) shows that the effective medium approximation reduces the dielectric parameters by a small amount. At higher droplet concentrations, this reduction becomes significant.

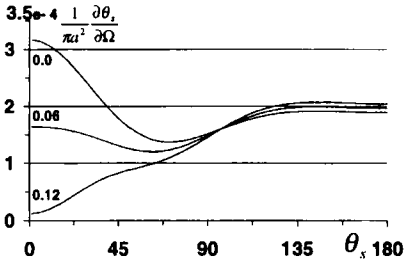
In figure 4(d), we show the effective differential cross-section using the hole structure factor and the effective medium approximation. The small reduction in the differential cross-section at the higher values of η follows from the reduction in the dielectric constants shown in figure 4(c).

Percus-Yevick hard spheres structure factor

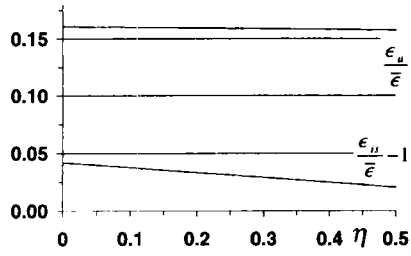
Figure 4(e) shows the differential cross-section using only the Percus-Yevick hard-spheres approximation, we may now consider higher droplet concentrations than previously. The affect of the different structure factor– shown in figure 4(e)– can be understood from figure 3. For large values of θ_s (the back-scattering régime) the structure factors, and hence the effective differential cross-sections, are similar. For forward scattering the Percus-Yevick structure factor is larger than the hole approximation adopted by Žumer *et al* [8] at a given η and hence the Percus-Yevick structure factor predicts a larger effective differential cross-section.



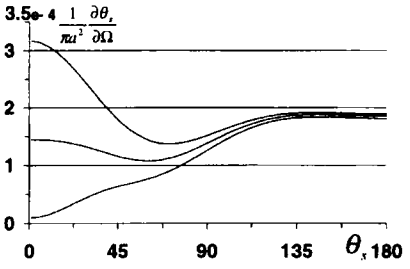
(a) Hole approximation (rounded dielectric parameters)



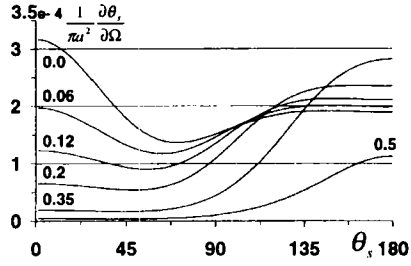
(b) Hole Approximation (exact dielectric parameters)



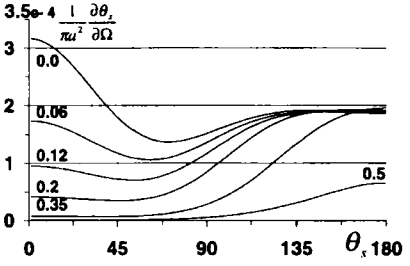
(c) Effective Medium Approximation



(d) Hole approximation including effective medium approximation



(e) Percus-Yevick (hard spheres)



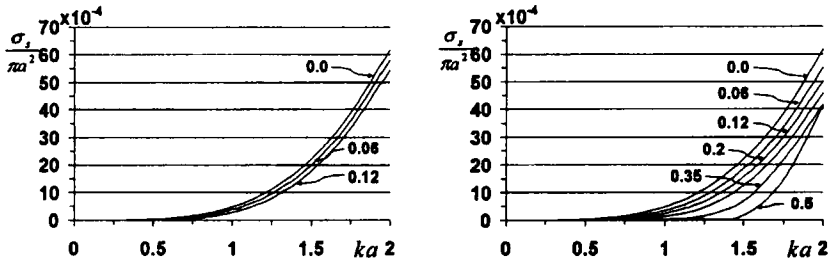
(f) Percus-Yevick and effective medium approximation

FIGURE 4 Effective differential cross-section versus scattering angle for droplets with a radial structure field for ka =1.5, for a variety of values of the packing fraction, η. See text for further details.

Finally, in figure 4(f), we combine both the Percus-Yevick structure factor. The effective medium approximation reduces the scattering at higher values of η. In practice, the angular dependence of the differential cross-section may differ from theoretical predictions due to, for example, multiple scattering, size inhomogeneities and our assumption that the droplets are homogenous. In principle we could introduce an effective droplet size and effective droplet pinning to address some of these problems.

Scattering cross-section, σ_s

In figure 5 we compare the scattering cross-section, σ_s, calculated as in table I, using our full effective medium calculation and Percus-Yevick structure factor with that obtained using the hole approximation.



(a) Hole approximation

(b) Percus-Yevick structure factor and effective medium approximation

FIGURE 5 Total scattering cross-section for radial droplets.

Figure 5 clearly shows a $(ka)^4$ dependence, characteristic of Rayleigh scattering. More interesting is the dependence of the scattering on the values of η shown. At low η, there is little difference between the two approaches. As η increases above 0.125, the hole approximation becomes unphysical, but using the Percus-Yevick theory allows us to investigate higher droplet

concentrations. The decrease in the total scattering as η increases is due to destructive interference between correlated scatterers.

Summary of radial droplets

Using the hole approximation, our formalism reproduces the results of Žumer *et al* [8]. The hole approximation overestimates the effective differential cross-section for small values of the scattering angle compared to the Percus-Yevick theory. At low droplet concentrations, the effective medium makes only a small difference to the calculation, however at higher concentrations it must be included. In a system of radial droplets, there is only coherent scattering, and increasing the droplet concentration reduces the scattering, due to density superpositional destructive interference. Thus at high values of η radial droplets are of limited use due to the difficulty of arranging an optical contrast between off and on states.

Partially Ordered Bipolar System

We now turn our attention to the technologically important case of the partially ordered bipolar system, shown schematically in figure 1. We have discussed the theoretical details of this system elsewhere[4, 5, 6], but repeat key formulae here. Finally we will show that a PDLC device based on partially ordered bipolar droplet has the properties which we require: it is opaque in the off state, and transparent in the on state. We separate the contribution from the coherent and incoherent scattering to understand the behaviour of the system in the off and on states, and show the transition between these states.

Coherent and incoherent scattering formulae

In Table II, we summarise the formulae required to calculate the scattering in the partially ordered system. In the case of the partially ordered system, there is a contribution from incoherent scattering, caused by uncorrelated scattering from droplets with different alignments to the field.

TABLE II Coherent (I_c) and incoherent (I_I) scattering formulae for partially ordered system.

$$I_C(\theta_s) = \frac{a^6 k^4}{18}[f(qa)]^2 \left[\frac{(\epsilon_\perp - \epsilon_m) + \frac{1}{3}(1-QS)(\epsilon_{11} - \epsilon_\perp)}{\epsilon_m}\right]^2 (2 - \sin^2(\theta_s))$$

$$I_I(\theta_s) = \frac{a^6 k^4}{8}[f(qa)]^2 \left[\frac{2S(\epsilon_{11} - \epsilon_\perp)}{3\epsilon_m}\right]^2 (c_1 \sin^2(\theta_s) + c_2)$$

$$S = 12\ln 2 - \frac{15}{2} \approx 0.818.$$

$$f(qa) = 3\frac{\sin(qa) - qa\cos(qa)}{(qa)^3}$$

$$c_1 = 2c_{xz} - c_{xx} - c_{xy}$$

$$c_2 = 2(c_{xx} + c_{xy})$$

$$c_{xz} = \frac{7 + 5Q - 12Q_4}{105}$$

$$c_{xy} = \frac{7 - 10Q + 3Q_4}{105}$$

$$c_{xx} = \frac{28 - 20Q - 35Q^2 + 27Q_4}{315}$$

S is an order parameter which describes the director distribution inside a bipolar droplet [6, 21].

Effective Medium Theory

In the case of the partially ordered system, we adopt an interpolation formula, (7), to account for the changes in the dielectric properties of the effective medium, as the droplet concentration increases. Hence we replace ϵ_m in table II by $\bar{\epsilon}$:

$$\bar{\epsilon} = (1-\eta)\epsilon_m + \eta\left[\epsilon_\perp + \frac{1}{3}(1-QS)(\epsilon_{11} - \epsilon_\perp)\right] \qquad (9)$$

Intensity of light

At zero field, the droplets are randomly oriented. Figure 6 shows the dependence of the intensity of transmitted light on the film thickness at zero field. Our theory agrees well with the experimental results of Žumer *et al* [8], we predict that the attenuation coefficient (Table I) $\mu = 3.8\times10^{-3}\mu m$, compared to the experimentally observed value of $\mu = 3.9\times10^{-3}\mu m$.

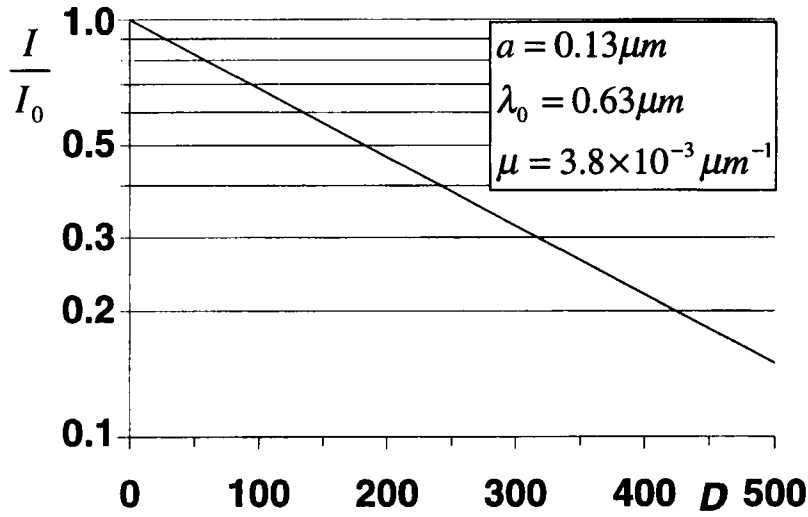


FIGURE 6 Intensity of light versus film thickness, D, for the partially ordered system in zero field.

Field dependence of attenuation coefficient

In figure 7, we use our full theory and values of Q calculated numerically[4] to derive the field dependence of the attenuation coefficient. The scaling of the applied voltage, $\tilde{U}$, is dependence on the system characteristics. For a switching transition at around 15 volts, film thickness, $D \sim 100\mu m$, and $a \sim 0.13\ \mu m$, we can estimate that $W \approx 4.7\times10^{-2}$ erg cm^{-2}. Further experimental work is required to investigate this parameter.

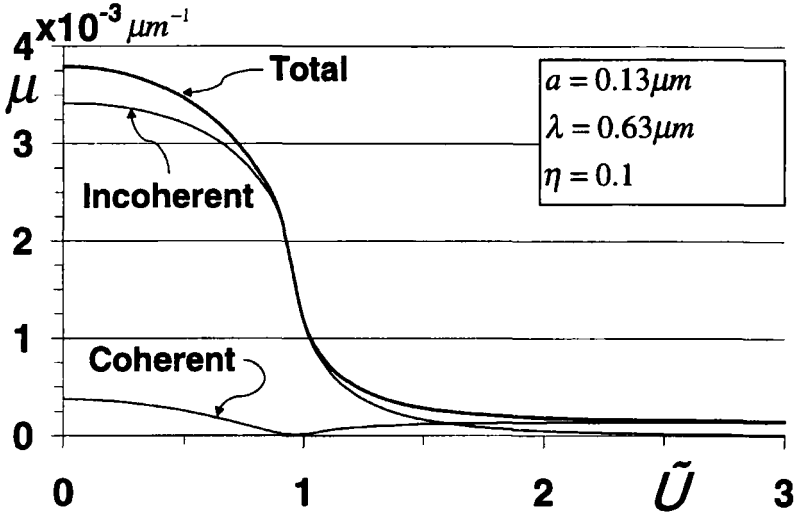


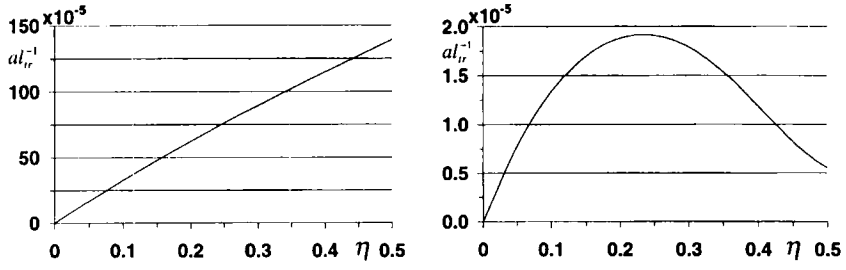
FIGURE 7 Attenuation constant, μ, versus scaled applied voltage for the partially ordered system.

At low fields, the scattering is predominantly incoherent, caused by the random alignment of the droplets. At high fields, the incoherent scattering vanishes identically, and the only contribution to the scattering is coherent.

Effect of droplet concentration on scattering

To design a PDLC, we require that it should be translucent in the off state, and transparent in the on state. As shown in figure 7, at low fields, the scattering is predominantly incoherent, and at high fields it is predominantly coherent. Since the incoherent scattering is caused by uncorrelated orientational fluctuations in the medium, the scattering is additive. Thus we may increase the scattering at low field by simply increasing the droplet concentration, η; this ensures that the PDLC is translucent at low fields. At high fields, the scattering is mainly coherent. In this case, increasing the droplet concentration decreases the scattering due to density superpositional interference. The PDLC is thus transparent at high fields! Figure 8 shows the

effect of the droplet concentration, η, on the inverse transport mean free path at low and high fields.



(a) Low Field ($\tilde{U} = 0$) (b) High Field ($\tilde{U} = 5$)

FIGURE 8 Inverse transport mean free path versus droplet concentration, η at low and high fields.

For the partially ordered droplets, we can ensure a high constrast ratio by simply increasing the droplet concentration.

DISCUSSION AND CONCLUSIONS

In this paper we have presented results for the theory of light scattering in the long wavelength régime where the Rayleigh-Gans approximation applies. We have made a detailed comparison between the predictions of our approach, developed elsewhere[4,5,6], and the theory developed by Žumer *et al*[8]. Our approach develops the work of Žumer *et al*[8] and Kelly and Palffy-Muhoray[9].

Our theory takes into account droplet correlations and the droplet concentration. We have demonstrated quantitatively that our basic theory reproduces exactly simple cases studied by Žumer *et al*[8], in the limit where our corrections are removed. We can understand qualitatively the enhancements from our approach in terms of our modified structure factor and effective medium theory. This approach is applicable to more general

problems in light scattering in inhomogeneous materials. For the partially ordered system, we have agreement with experimental results for low fields, and can describe the field dependence of the light scattering in the film.

In the future we will extend our approach to incorporate light scattering for large values of *ka* (the so called anomalous diffraction régime) and non-normal incidence of light onto the PDLC film.

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