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Light scattering from polymer-dispersed liquid crystal films: Droplet size effects

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The effects of droplet size on light scattering by polymer-dispersed liquid crystal (PDLC) films have been studied theoretically and experimentally. Rayleigh–Gans calculations predict that, provided the droplets scatter independently, backscattering will be maximum at wavelength \(\lambda\) when the droplet radius is about \(\lambda/7\). However, correlation effects become noticeable for droplet volume fractions \(\eta\) above about 0.2 and shift the radius for maximum backscattering to about \(\lambda/5\) as \(\eta\) increases to 0.5. These calculations are consistent with hemispherical transmittance and reflectance measurements on PDLC films in which multiple scattering is known to be important. These measurements indicate that solar attenuation by PDLC films will probably be maximized by choosing droplet size to maximize backscattering of visible radiation. This choice also increases solar attenuation by absorption of radiation trapped inside a PDLC film by total internal reflection.

I. INTRODUCTION

Polymer-dispersed liquid crystal (PDLC) films, comprised of liquid crystal (LC) microdroplets dispersed in polymer matrices, are potentially useful for solar energy control and other electro-optic applications because they can be switched electrically from a cloudy, light-scattering state to a transparent state.\(^\text{1-3}\) Optimizing the light scattering performance of a PDLC film for a specific application requires control over various film parameters including the refractive indices of the liquid crystal and polymer matrix materials and the concentration and size distribution of the liquid crystal droplets in the film. In this paper we describe the effects of droplet size on light scattering by PDLC films with particular emphasis on maximizing backscattering for solar energy control.

We first use the Rayleigh–Gans (RG) approximation\(^\text{4-6}\) to study the effect of droplet size on the energy backscattered from and transmitted through a PDLC film (Sec. II). Our calculations predict an optimum droplet size for maximizing backscattering and minimizing energy transmission at a given wavelength. We then test these predictions by comparing them with measured light scattering properties of PDLC films with controlled droplet size distributions (Sec. III). Finally, we use these measurements to assess the effect of droplet size on solar attenuation by PDLC films (Sec. IV).

II. RAYLEIGH–GANS ANALYSIS OF DROPLET SIZE EFFECTS IN LIGHT SCATTERING FROM PDLC FILMS

A. Analytical procedure

An exact analysis of light scattering by PDLC films is not possible because no theory of multiple scattering by a system of optically anisotropic particles has yet been developed. However, light scattering by PDLC films is predominantly forward scattering.\(^\text{7}\) Consequently, even though multiple scattering is important in these films,\(^\text{7}\) most of the light initially scattered into the backward hemisphere by a droplet will continue to propagate into the backward hemisphere after successive scatterings and the net backscattering from the system should be maximized by choosing the droplet size to maximize backscattering from a single droplet. Therefore, to understand the influence of droplet size on backscattering, we will neglect multiple scattering effects and consider single scattering by a collection of LC droplets.

In a PDLC film, each droplet has an axis of symmetry (optic axis) determined by the orientations of the optically anisotropic LC molecules within the droplet. Light polarized parallel to the optic axis will be scattered differently from light polarized perpendicular to this axis. As a result, the differential scattering cross section of a LC droplet will depend on its orientation relative to the propagation direction of the incident light. In a PDLC film in its off state, the orientation of the optic axis varies randomly from droplet to droplet. Thus, scattering from a collection of LC microdroplets is considerably more complicated than scattering by a collection of optically isotropic particles, even in the single-scattering approximation.

Zumer and Doane\(^\text{8}\) have recently extended the Rayleigh–Gans (RG) scattering theory to describe scattering by a single nematic LC droplet. In general, calculation of the differential scattering cross section of a droplet requires numerical integration over the spatially nonuniform refractive index distribution within the droplet. Clearly, the effect of optical anisotropy of the LC molecules on scattering would be greatest if all the molecular axes within the droplet were parallel to each other. In this case, the numerical integrations can be performed in closed form\(^\text{8}\) with the result that, regardless of the orientation of the droplet symmetry axis, the expression for the differential...
The scattering cross section for unpolarized light has exactly the same dependence on the droplet radius as in the isotropic case.\textsuperscript{4,6} The effect of anisotropy is to replace $|m - 1|^2(1 + \cos^2 \theta)$, which occurs in the formula(s) for the scattering cross section(s) of an optically isotropic droplet [see Eqs. (1)–(4) below], by an expression of the form $A(n_r, n_0) + B(\theta, n_r, n_0)$ where $A$ and $B$ are functions of $n_r$ and $n_0$, the extraordinary and ordinary refractive indices of the liquid crystal. Thus, the effect of maximum anisotropy is to change the magnitude of light scattering but not its size dependence. Since the differential cross section has the same size dependence in the limits of no anisotropy and maximum anisotropy, it seems quite reasonable to neglect effects of anisotropy altogether and use the simpler formulas for RG scattering by isotropic droplets to analyze droplet size effects. This is the approach we have taken in this work.

The validity of neglecting multiple scattering and anisotropy can be confirmed only by comparing results of our analysis with experiment. We do this in Sec. III.

In the absence of multiple scattering, the fraction $B$ of the incident power which is scattered out of a light beam into the backward hemisphere by a collection of identical droplets is given by:

$$B = N_o \sigma_d a,$$

where $N_o$ is the number of droplets per unit volume, $\sigma_d$ is the total backscattering cross section for a single droplet, and $d$ is the distance light would travel in the sample in the absence of scattering. For normally incident light, the only situation considered in this paper, $d$ equals the sample thickness. $N_o$ depends on the radius $a$ of each droplet and the volume fraction $\eta$ occupied by the droplets:

$$N_o = \frac{3 \eta}{(4\pi a^3)^{-1}}.$$  

(2)

Following the procedures of Ref. 6, we can show that, in the RG approximation, the backscattering cross section of a single optically isotropic droplet may be written for unpolarized light as:

$$\sigma_d = \frac{\pi a^2}{|m - 1|^2} \Phi_b(ka),$$  

where

$$\Phi_b(ka) = \frac{4}{9} (ka)^2 \int_0^\pi (1 + \cos^2 \theta)$$

$$\times \left[ \frac{3}{(ka)^2} (\sin k_a a - k_a a \cos k_a a) \right]^2$$

$$\times [1 + \beta N_o \hat{h} (k_a, \eta, a)] \sin \theta d\theta,$$  

(4)

In Eqs. (2)–(4), $k = 2\pi n_f / \lambda$ is the magnitude of the wavenumber of the incoming radiation inside the polymer matrix; $\lambda$ is the vacuum wavelength of this radiation, $m = n_{LC}/n_0$ is the ratio of the liquid crystal refractive index $n_{LC}$ to the polymer matrix refractive index $n_0$, and $k$ is related to the scattering angle $\theta$ between the propagation directions of the incident and scattered light by $k = 2k \sin(\theta/2)$.

Figure 1 shows the function $\Phi_b(ka)/ka$ as a function of $ka$ (lower scale) and $a/\lambda$ for $n_f = 1.52$ (upper scale). The curve labeled “No Correlation” is for independent scattering and does not depend on volume fraction. The other curves, which depend on volume fraction, include effects of correlation on the scattering.

The factor $\beta$ in Eq. (4) is 0 if the droplets scatter independently and 1 if correlation effects are important. Correlation effects have been discussed in considerable detail in Ref. 6, in which the Percus–Yevick approximation\textsuperscript{6} for the radial distribution function of a system of hard spheres was used to derive an explicit expression for $\hat{h} (k_a, \eta, a)$, the function which describes the effect of correlation. We shall use these results in our calculations for $\beta = 1$.

Combining Eqs. (1)–(4), we obtain

$$R = N_o \sigma_d a = \frac{3\pi n_f d}{2 \lambda} \left[ \frac{m - 1}{\eta} \right] \frac{\Phi_b(ka)}{ka}.$$  

(5)

Finally, we note that $T$, the fraction of the incident power which is transmitted into the forward hemisphere, is the sum of the (fractional) power which is transmitted without scattering and the (fractional) power which is scattered into the forward hemisphere. In a nonabsorbing system, energy conservation requires that

$$T = 1 - B.$$  

(6)

B. Results

The validity of neglecting multiple scattering and anisotropy can be confirmed only by comparing results of our analysis with experiment. We do this in Sec. III.

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(6)

B. Results

Figure 1 shows the function $\Phi_b(ka)/ka$ as a function of $ka$ (lower scale) and $a/\lambda$ for $n_f = 1.52$ (upper scale). For independent scattering (no correlation), $\Phi_b(ka)/ka$ does not depend on volume fraction and is shown by the solid curve. The other curves include the effects of correlation and depend on volume fraction. When correlation effects are included, the value of $a/\lambda$ that maximizes $\Phi_b(ka)/ka$ increases with increasing volume fraction $\eta$ for $0 < \eta < 0.5$; however, the maximum value of $\Phi_b(ka)/ka$ does not vary significantly with $\eta$ until $\eta$ exceeds 0.4.

To determine the fractional powers $B$ and $T$, we must specify the wavelength $\lambda$, the film thickness $d$, the relative refractive index $m$ and the volume fraction $\eta$ in Eq. (5). We shall set $m = 1.0428$ (i.e., $n_{LC} = 1.585$) and $d = 4 \mu$m.
FIG. 2. Fractional transmitted power $T = 1 - B$ vs droplet radius $a$ for $\lambda = 632.8$ nm. (a) independent scattering, (b) with droplet correlation.

In the absence of correlation ($\beta = 0$), the fractional backscattered power $B$ is directly proportional to the volume fraction occupied by the liquid crystal droplets and the fractional transmitted power $T = 1 - B$ will decrease linearly with increasing volume fraction. At any wavelength $\lambda$, backscattering is a maximum and transmitted power a minimum when the droplet radius $a \approx \lambda/7$. These results are illustrated in Fig. 2(a), which shows the fractional transmitted power as a function of droplet radius $a$ for $\lambda = 632.8$ nm.

Figure 2(b) shows the fractional transmitted power $T$ as a function of $a$ for $\lambda = 632.8$ nm when correlation effects are included in the calculations ($\beta = 1$). At low volume fractions ($\eta < 0.2$), the curves are only slightly different from those obtained when correlation effects are neglected. At higher volume fractions, however, the curves differ noticeably from those obtained in the absence of correlation.

The dependence of the fractional transmitted power $T$ on volume fraction $\eta$ is very complicated because, when correlation effects are important, the linear dependence of $B$ on $\eta$ is modified by the complex dependence of $\Phi_b(ka)/ka$ on $\eta$ (see Fig. 1). The key characteristics of backscattering including correlation effects may be summarized as follows.

1. The fractional backscattered power $B$ is always $< 1$ its value in the absence of correlation so $T$ is always $> 1$ its value for independent scattering. (2) $B$ no longer increases monotonically with increasing $\eta$ but reaches a maximum value when $\eta$ lies near 0.5. (3) As $\eta$ increases to 0.5, the droplet radius which maximizes $B$ and minimizes $T$ increases from $\lambda/7$ to about $\lambda/5$.

We have also confirmed by direct calculation that, for a fixed droplet size, $B$ is reduced and $T$ increases as wavelength increases. Thus, even with droplet size optimization, it will be very difficult to make PDLC films that backscatter efficiently at infrared wavelengths.

Although our primary interest in this paper is understanding the relationships between droplet size and backscattering, we have also calculated the effects of droplet size and concentration on $S$, the total fraction of the incoming power which is scattered out of a beam. These effects may be important in PDLC applications in which minimizing off-state transmittance in the direction of the incident beam is important, e.g., in projection displays. $S$ may be computed from Eq. (5) by replacing $\Phi_b(ka)$ by $\Phi(ka)$, which is obtained by changing the lower limit of integration from $n/2$ to 0 in Eq. (4).

Results of such a calculation are shown in Fig. 3 for the same parameters used in calculating the curves of Fig. 2. For a fixed volume fraction $\eta$, $S$ increases monotonically with increasing $a/\lambda$ with or without correlation. Correlation significantly influences the effect of $\eta$ on $S$. For a fixed radius, $S$ initially increases with increasing $\eta$, reaches a maximum value for $\eta$ between 0.3 and 0.4, and then decreases. This behavior results from two competing effects:
$S$ is directly proportional to $\eta$ [Eq. (5)]; however, $\Phi(ka)/ka$ decreases as $\eta$ decreases. At low concentrations, the linear dependence on $\eta$ dominates and $S$ increases with increasing $\eta$. At higher $\eta$ values, the decrease in $\Phi(ka)/ka$ dominates and further increases in $\eta$ lead to decreased scattering. This behavior differs from that obtained in the absence of correlation. In that case, $S$ increases linearly with $\eta$ since $\Phi(ka)/ka$ is independent of $\eta$.

**III. EXPERIMENTAL VERIFICATION OF LIGHT SCATTERING ANALYSIS**

The analytical results presented in Sec. II indicate that droplet radii small compared to the incident wavelength are needed to maximize backscattering. To test the theoretical predictions, we have measured the transmitted and reflected powers from thermally cured samples with controlled droplet size distributions and compared the experimental data with our calculations.

**A. Samples**

Five samples were studied. All utilized 26 $\mu$m diam spacers to obtain uniform film thickness. The polymer matrix in each sample was formed from Capcure 3-800 hardener and an equivalent weight of a resin consisting of 65 wt. % MK-107 and 35 wt. % Epon 828. Each sample contained 41 wt. % of the commercial LC mixture E7. To obtain reliable estimates of the droplet sizes in our samples we used results of extensive scanning electron microscopy (SEM) studies which relate droplet size to cure temperature for PDLC films having the same composition as those used in the present study.

It is well known that the polymerization rate $1/\tau$ depends exponentially on temperature:

$$\frac{1}{\tau} \sim e^{-E_{\text{act}}/kT},$$

where $E_{\text{act}}$ is an activation energy, $k$ is Boltzmann’s constant, $\tau$ is the cure time constant, and $T$ is the absolute temperature. Furthermore, studies in several laboratories have shown that droplet size increases as the cure time constant increases. The functional form of the dependence of droplet size on time constant has not been determined precisely; however, if the (average) droplet radius $\bar{a}$ were proportional to some power of the cure time constant, Eq. (7) would give

$$\bar{a} \propto \alpha^B e^{E_{\text{act}}/kT},$$

where $A$ and $B$ are constants. In this case, a plot of $\ln \alpha$ vs $1/\tau$ would be a straight line. Figure 4(a) shows a plot of $\ln \alpha$ vs $1000/T$ for samples with the same composition as those used in our measurements. The circles are droplet radii determined by scanning electron microscopy; the solid line is a linear least-squares fit to the SEM data, confirming the dependence of size on temperature.

The fraction of the total area occupied by droplet cross TABLE I. Characteristics of PDLC samples. In all samples the polymer matrix was formed from a mixture containing 65% MK-107, 35% Epon 828, and an equivalent weight of Capcure 3-800. Each sample was made with 26 $\mu$m spacers. Radii and volume fractions were determined from regression analysis of scanning electron micrograph data.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Wt. %</th>
<th>Cure temperature (°C)</th>
<th>Radius (fit) (µm)</th>
<th>Volume fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. 1</td>
<td>41</td>
<td>50</td>
<td>3.60</td>
<td>0.19</td>
</tr>
<tr>
<td>No. 2</td>
<td>41</td>
<td>60</td>
<td>1.77</td>
<td>0.14</td>
</tr>
<tr>
<td>No. 3</td>
<td>41</td>
<td>70</td>
<td>0.91</td>
<td>0.10</td>
</tr>
<tr>
<td>No. 4</td>
<td>41</td>
<td>80</td>
<td>0.48</td>
<td>0.07</td>
</tr>
<tr>
<td>No. 5</td>
<td>41</td>
<td>100</td>
<td>0.15</td>
<td>0.04</td>
</tr>
</tbody>
</table>

The good agreement between the calculated and measured radii allows us to use this curve to determine the droplet radii in our samples. The results are shown in Table I, together with other sample characteristics discussed earlier. We emphasize that, while Eq. (8) is satisfied for the samples described in this paper and provides a convenient way for us to relate droplet size to cure temperature, there is no a priori reason to expect it to hold in general.

The fraction of the total area occupied by droplet cross...
sections on an SEM micrograph of a cut through a PDLC film is the product of the number of droplets per unit area and the (average) cross-sectional area of a droplet. This fractional area is equal to the volume fraction \( \eta \) occupied by the droplets in the film.\(^{14} \) If all the droplets are approximately the same size, Eq. (8) implies that \( \ln \eta \) is a linear function of \( 1/T \). Therefore, linear regression analysis of experimental SEM area fraction data can be used to relate \( \eta \) to cure temperature. The volume fractions given in Table I were obtained from such an analysis. The decrease in volume fraction with increasing cure temperature in epoxy-based PDLC films has been verified by calorimetry.\(^{12} \)

The volume fractions of Table I are substantially lower than the initial LC concentrations in the mixtures from which the films were formed. Calorimetric and SEM studies\(^{2,12,13} \) have shown that a significant fraction of liquid crystal may remain dissolved in the matrix upon polymerization; this residual LC acts as a plasticizer. The amount of liquid crystal which remains dissolved depends on the particular materials used and on the degree of cross linking and appears particularly large in the PDLC system described in this paper. We believe that this accounts for the very large differences between the initial LC concentrations in the starting mixtures and the volume fractions in the droplets in the cured films.

**B. Measurements of fractional reflected powers**

Using the integrating sphere accessory on a Perkin-Elmer \( \lambda \)-9 spectrophotometer, we measured the wavelength-dependent hemispherical reflectance of each PDLC sample described in Table I. The power which is specularly reflected from the sample surfaces and the power which is scattered into the backward hemisphere by the LC microdroplets both contribute to the measured reflectance. The results, plotted in Fig. 5, demonstrate clearly that backscattering increases monotonically with decreasing droplet size for all the droplet radii we studied and that most of the backscattering occurs at visible wavelengths.

![FIG. 5. Hemispherical reflectance vs wavelength for PDLC samples. The reflectance, measured with an integrating sphere, includes both backscattered and specularly reflected radiation. The curve for each sample is labeled by the droplet radius in the sample.](image1)

![FIG. 6. Hemispherical reflectance vs droplet size for PDLC samples. The values at 633 and 1500 nm were obtained from the spectra of Fig. (5); the solar values were obtained by weighting the spectra of Fig. (5) with air mass two solar irradiance values and integrating over appropriate wavelength regions.](image2)

**C. Comparison with theoretical predictions**

We used the spectral curves of Fig. 5 to determine the reflectance as a function of droplet size at selected wavelengths. Representative results are shown in Fig. 6. Each open circle is the reflectance of one PDLC sample at 633 nm; the reflectances are plotted as a function of the droplet radius in the sample. At this wavelength, reflectance increases monotonically as the droplet size decreases. This behavior is consistent with our calculations which predict that, at 633 nm, maximum backscattering should occur for a droplet radius of 0.09 \( \mu \)m, which is slightly below the smallest radius in any of our samples. For the low droplet volume fractions in our samples, the predicted sizes for maximum backscattering do not depend on whether correlation effects were included in the calculations.

Our calculations also predict that maximum backscattering at 1500 nm should occur for a droplet radius of 0.22 \( \mu \)m. The open squares denote measured reflectances at this wavelength. The maximum reflectance is found in sample No. 4 in which the droplet radius is 0.48 \( \mu \)m. This reflectance is only a few tenths of a percent higher than that measured in sample No. 3 (\( \alpha = 0.91 \mu \)m) and in sample No. 5 (\( \alpha = 0.15 \mu \)m); however, these differences are not significant since the precision in our reflectance measurements is, at best, 1%. Therefore, although we cannot confirm precisely the predicted radius for maximum backscattering at 1500 nm, we can say that the predicted value is not inconsistent with our measurements.

Figure 7 shows the power scattered in the off-state by samples Nos. 2 and 5 as a function of the scattering angle \( \Theta_s \). These data were obtained with the upgraded version\(^{15} \) of the experimental system described in detail in Ref. 7. The system uses well-collimated light from a HeNe laser operating at 632.8 nm and, at each scattering angle, collects light within a cone of 0.5° half angle about the axis of

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**References**

the detection system. The curves in Fig. 7 show the power measured by the detector at each scattering angle normalized to the incident laser power.

When the scattering angle is zero, the system measures (essentially) the unscattered power which propagates through the sample in the direction of the incident laser beam. Approximately 0.5% of the incident power is transmitted through sample No. 2 without scattering whereas about 3.5% is transmitted through sample No. 5. These results are qualitatively consistent with our calculations which predict that $S$, the fraction of the incident power scattered out of an incoming light beam, increases with increasing droplet size.

The data of Fig. 7 also show that most of the scattered light from sample No. 2 enters the forward hemisphere whereas a much higher fraction of the incoming light is backscattered by sample No. 5. (In comparing the two curves of Fig. 7 one should remember that, at each scattering angle, the experimental system measures the signal received by a detector with a fixed small solid angle. As the magnitude of the scattering angle increases, this detector captures a smaller fraction of the total power scattered at this angle, which must be obtained by azimuthal integration.)

IV. EFFECT OF DROPLET SIZE ON SOLAR ATTENUATION BY PDLC FILMS

In addition to providing verification of our light scattering calculations, the data of Fig. 5 can be used, together with transmittance data presented below, to assess the solar attenuation characteristics of PDLC films.

A. Effect of droplet size on solar-weighted reflectances

We have weighted each spectrum in Fig. 5 with the air mass two (AM2) spectral distribution of solar irradiance at the earth's surface and integrated over the appropriate wavelength ranges to find the fractions of solar energy which would be reflected by each of our samples in the ultraviolet (UV), visible, and near infrared (NIR) portions of the solar spectrum. The results are summarized in Table II and plotted in Fig. 6 for comparison with the reflectances at 633 and 1500 nm. The solid squares in this figure are solar-weighted NIR reflectances. The maximum NIR reflectance was measured in sample No. 4 ($a=0.48 \mu m$). By contrast, the solar-weighted visible (solid circles) and total solar (triangles) reflectances increase monotonically with decreasing droplet radius for all the droplet radii that we studied experimentally. These data show that the backscattering of solar radiation is dominated by backscattering at visible wavelengths and suggest that solar attenuation by PDLC films will probably be maximized by choosing droplet size to maximize backscattering of visible solar radiation.

B. Fractional transmitted power: Importance of LC concentration

In Fig. 8 we show hemispherical transmittance spectra for each of the samples of Table I measured with the integrating sphere on our $\lambda$-9 spectrophotometer. Radiation which passes through the sample without scattering and radiation which is scattered out of the incoming beam into angles within the forward hemisphere both contribute to

<table>
<thead>
<tr>
<th>Sample</th>
<th>IUV (295-400 nm)</th>
<th>VIS (400-700 nm)</th>
<th>NIR (700-2140 nm)</th>
<th>Total (295-2140 nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Forward</td>
<td>Backward</td>
<td>Forward</td>
<td>Backward</td>
</tr>
<tr>
<td>No. 1</td>
<td>61.8</td>
<td>20.2</td>
<td>88.9</td>
<td>13.1</td>
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<tr>
<td>No. 2</td>
<td>47.4</td>
<td>27.3</td>
<td>75.1</td>
<td>16.4</td>
</tr>
<tr>
<td>No. 3</td>
<td>41.7</td>
<td>30.1</td>
<td>67.2</td>
<td>18.3</td>
</tr>
<tr>
<td>No. 4</td>
<td>39.0</td>
<td>31.0</td>
<td>62.3</td>
<td>19.9</td>
</tr>
<tr>
<td>No. 5</td>
<td>33.5</td>
<td>37.3</td>
<td>56.3</td>
<td>23.2</td>
</tr>
</tbody>
</table>
the measured transmittance. As expected from the reflectance spectra of Fig. 5, transmittance decreases with decreasing droplet radius. Solar-weighted transmittances for these samples are summarized in Table II.

The lowest transmittance measured in these samples may still be too high for some solar control applications. We attribute this to the low droplet concentrations in the samples (Table I and Sec. III A). Our calculations have shown that, at these low LC concentrations, correlation effects are negligible and the fractional scattered powers $S$ and $B$ increase linearly with the LC volume fraction $\eta$, as does the fractional forward scattered power $F$ which we have not discussed explicitly in this paper. For low $\eta$ values, all these scattered powers are small and much of the incident energy passes through the sample without scattering. Thus, the fractional transmitted power is high even in the samples in which the droplet size produces efficient backscattering. Clearly, efficient attenuation of solar radiation requires a high concentration (large volume fraction) of LC droplets of the proper size.

C. Effect of droplet size on internally reflected power

Inspection of the data in Table II shows that transmittance and reflectance do not usually add to unity even when measured with an integrating sphere. The reason for this has been discussed previously: scattered light which reaches an air-glass interface of a PDLC film will be totally internally reflected if its propagation direction makes an angle larger than the critical angle with the normal to the interface. Since the fraction of incident light which is trapped in a PDLC film by total internal reflection affects its solar control characteristics, it is useful to know how the fraction of trapped radiation depends on droplet size.

Figure 9 shows the percent of incident radiation trapped by internal reflection for each of our PDLC samples. These values were obtained by adding together the percent transmitted and reflected powers and subtracting from 100%. Recent measurements using calibrated reflectance standards indicate that our integrating sphere reflectances may be as much as 3% too high; this overestimate causes some of the computed trapped power values for sample No. 1 (droplet radius = 3.6 $\mu$m) to be less than zero.

We shall not attempt to relate trapped power to our scattering calculations. Computation of trapped power from scattering theory would require us to calculate the angular distribution of scattered radiation as a function of droplet size. We know that this cannot be done reliably using single scattering theory even if optical anisotropy is taken into account. Despite these limitations, the data of Fig. 9 show clearly that trapped power generally increases with decreasing droplet size for the range of droplet radii studied. In practice, most of this trapped power will be absorbed. We have found that, although absorption in PDLC films is usually negligible over distances comparable to the film thickness, it can be significant over distances of several centimeters and will certainly be important over distances comparable to the dimensions of windows, sunroofs, and similar systems.

V. SUMMARY AND CONCLUSIONS

We have studied, both theoretically and experimentally, the effects of droplet size on the scattering of light by PDLC films. Our theoretical analysis was based on the Rayleigh–Gans approximation in the single scattering regime with and without the effects of droplet correlation. Results of the calculations were compared with measurements on thermally cured PDLC films in which multiple scattering was known to occur. Despite the obvious limitations of single scattering analysis, many of its predictions were confirmed by, or at least found to be not inconsistent with, our experimental data. From this analysis and from extensive angle-dependent and angle-integrated light scattering measurements such as those illustrated in this paper,
we can summarize the major effects of droplet size on light scattering by PDLC films.

For droplets with radii much smaller than the incident wavelength, the total fractional scattered power $S$ is small. Most of the incoming radiation passes through the film without scattering. Half of the scattered radiation enters the forward hemisphere, and half enters the backward hemisphere ($F = B = S/2$).

As droplet size increases, $S$, $F$, and $B$ all increase. The ratio $F/B$ also increases. This behavior continues until the droplet size reaches the value which maximizes the fractional backscattered power $B$ at the incident wavelength. Our calculations show that this droplet radius is about $\lambda/2$ for droplet volume fractions below about 0.2. This theoretical result is consistent with our experimental data. If correlation effects are unimportant, this relation between radius and wavelength should hold for higher droplet concentrations as well; if correlation is important, however, the radius for maximum backscatter should increase to about $\lambda/5$ as the droplet volume fraction increases to 0.5. We have not yet studied scattering for high enough concentrations of small droplets to confirm these predictions experimentally.

As the droplet radius increases further, $B$ decreases while $S$ and $F$ increase. When the droplet size becomes very large, almost all the incoming radiation is scattered, only a small component is transmitted through the film without scattering, and almost all the scattering from each drop is forward scattering.

Comparison of these predictions with our experimental data indicates that the main effects of multiple scattering are to increase the overall level of scattering beyond that predicted by single scattering theory and to increase the fraction of backscattering by repeated interaction of forward-scattered light with additional droplets.

We have also measured the fractional forward-scattered and backscattered powers from our samples as a function of wavelength and used the results to assess the solar attenuation characteristics of PDLC films. We found that backscattering of solar radiation is dominated by scattering at visible wavelengths. Consequently, solar attenuation by PDLC films will probably be maximized by choosing droplet size to maximize visible backscattering. Whether a distribution of droplet sizes or a nearly monodisperse system of droplets is preferable is still to be determined. It will be very difficult to obtain efficient backscattering at infrared wavelengths even with droplet size control.

Our data indicate that the fraction of the incoming power trapped inside a PDLC film by total internal reflection increases as the droplet radius decreases. In a film with large area, most of this trapped radiation would be absorbed. Thus, controlling droplet size to maximize backscattering also increases solar attenuation by absorption.

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10. E7 is a commercial mixture of biphenyl and terphenyl liquid crystal compounds available from EM Chemicals, Hawthorne, NY.