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Angular discrimination of light transmission through polymer-dispersed liquid-crystal films

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The refractive index of the polymer n_p is adjusted relative to the ordinary refractive index of dispersed droplets of nematic liquid crystal n_o to regulate the angle of maximum transmission through a film in the presence of an applied field. Maximum transmission occurs at normal incidence when $n_p < n_o$ with the breadth of the angle of view being largest at $n_p = n_o$. When $n_p > n_o$, maximum transmission is peaked at an angle away from normal incidence, the value of the angle depending upon the ratio n_p/n_o . Angular discriminating filters are therefore possible with these films. The optical response of these materials as a function of applied field is also shown to be dependent on the ratio n_p/n_o as is the contrast exhibited by information displays. Methods for measuring the scattering cross section of a nematic droplet are also described.

I. INTRODUCTION

Nematic liquid crystals dispersed as micrometer-size droplets in polymer films have been of recent interest as optically switchable coatings and films for use in information displays or other devices.¹⁻⁵ In previous communications, only the index-matched condition, $n_p/n_o = 1$, has been discussed where n_p is the refractive index of the polymer and n_o the ordinary refractive index of the liquid crystal.¹⁻⁵ In this condition light transmission through a film in an applied field of sufficient strength to align the nematic droplets is maximum at normal incidence to the film's surface. We shall show that the angular breadth of the transmission peak is also broadest in this matched case. It is found, however, that the transmission peak narrows while remaining centered at normal incidence as the ratio n_p/n_o becomes less than one. For ratios of $n_p/n_o > 1$ the transmission peak is shifted away from normal incidence with the angle of maximum transmission depending on the value of the ratio n_p/n_o .

These features are explained in terms of effective index n_d for a nematic droplet and the dependence of the scattering of polarized light on the difference $(n_p - n_d)$. Measurements are performed on four samples: $n_p/n_o = 0.993$, $n_p/n_o = 1.006$, $n_p/n_o = 1.019$, and $n_p/n_o = 1.046$ made by phase separation methods using different polymer materials. These measurements illustrate the effectiveness of polymer dispersed liquid crystals (PDLCs) as angular discriminating filters. From these measurements, the effect of film thickness on light transmission is also determined. The dependence is found to be exponential in the range studied, and a value of the product of the scattering cross section of droplet and the droplet density is measured.

Finally, it is known that the sign of $(n_p - n_o)$ can be determined by the dependence of transmitted light on the voltage applied to a sample cell. Distinctive features in the "threshold" curve indicate whether the difference $(n_p - n_o)$ is positive, negative, or near zero.

II. MATERIALS AND SAMPLES

Light valves are prepared from polymer-dispersed liquid crystal (PDLC) materials by one of two methods: poly-

merization-induced phase separation (PIPS)^{2,4} and thermal-induced phase separation (TIPS).⁶ In PIPS thermoset polymer precursors are mixed with the liquid crystal to form a homogeneous solution. The solution is sandwiched between transparent conducting substrates with glass spacers used to control the thickness. The material is then cured at the appropriate temperature resulting in formation of the PDLC material. Light valves are also constructed by dissolving a thermoplastic and liquid crystal in a common solvent. The solution is poured on a transparent conducting substrate and the solvent evaporated resulting in formation of the PDLC film. A second substrate is placed on top of the PDLC followed by application of pressure and heat. This forms a sandwich in which the thickness is controlled by glass spacers. TIPS is then used to create droplets of specified dimension by controlling the rate of cooling from solution into the miscibility gap. A wide range of polymers may be employed in PDLCs making a wide range available for values of n_p .

Four different PDLC light valves were formed each with a different ratio of n_p/n_o (see Table I). The eutectic liquid-crystal mixture E7 (obtained from EM Chemicals) was used in all cases: $n_o = 1.520$, $n_e = 1.745$ at 20 °C.

The liquid crystal is dissolved to some extent in all of the cured polymers. This will influence the effective value of n_p depending on the amount of E7 present in the polymer. Not all components of E7 are likely to be equally dissolved in the polymer. The concentration of E7 in the polyvinylformal and polycarbonate materials is substantially greater than in the Epon systems. Isotropic E7 has a refractive index of about 1.59. Thus, the polycarbonate material will have a value of n_p somewhere between 1.585 and 1.59, still substantially greater than n_o . Likewise, the polyvinylformal material has a value of n_p somewhere between 1.502 and 1.59. Scanning electron microphotographs of the polyvinylformal films show that approximately 50% of the material is liquid-crystal droplets and 50% is the polymer binder. Thus, the polymer binder is composed of about 34% E7 and 66% polyvinylformal. Assuming the refractive index of the mixture is simply the average by weight of the relative contributions of

TABLE I. PDLC samples used in this study where the liquid-crystal material E7 (EM Chemicals) was used; $n_o = 1.520$, $n_e = 1.745$ at 20 °C.

Index match	Polymer	Refractive index			wt. % E7	n_p/n_o
		Pure ^a	Cured ^b	PDLC binder ^c		
$n_p > n_o$	polycarbonate	1.585	1.585	1.59	67	1.046
$n_p > n_o$	Epon 828 + Capcure 3-800	1.574 1.503	1.554	1.55	33	1.020
$n_p \approx n_o$	polyvinylformal	1.502	1.502	1.53	67	1.006
$n_p < n_o$	Epon 812 + Capcure 3-800	1.478 1.503	1.506	1.51	33	0.993

^aRefractive index of the pure materials.

^bRefractive index of the cured binder without dissolved liquid-crystal material.

^cRefractive index of the cured binder with dissolved liquid-crystal material.

the parts, the refractive index of the cured binder with dissolved liquid crystal is near 1.53. It will be shown that this value is close to the effective value of n_o of the droplet. This is in contrast to Epon 812/Capcure and Epon 828/Capcure which has little E7 dissolved and thus has an n_p essentially equal to that of the pure solidified polymer.

The liquid crystal in the droplets appears to be only weakly contaminated as the nematic-isotropic (NI) transition temperature is only slightly depressed ($\sim 2-4$ °C) from that of bulk E7. In the Epon samples the NI transition slowly improves with time, indicating droplet purification. We have found that dissolving 2 wt. % of epoxy into E7 will lower the transition by 4 °C. This may be compared with a detailed study of epoxy systems by differential scanning calorimetry which determined impurity levels in the range of 0.5 mol %.⁷

III. EFFECTIVE REFRACTIVE INDEX

The light scattering properties and hence the transparency of a PDLC film is a complex subject to describe theoretically and has only been worked out in the case of a single, submicrometer-size nematic droplet under a Rayleigh-Gans approximation.⁸ A simple and useful approach, though not as precise, uses the concept of an effective refractive index n_d for a liquid-crystal droplet. Such an approach is particularly useful to predict the transparency of a film for some applications. In the case of an aligned nematic, the effective index

n_α for the polarization indicated in Fig. 1 is given by the equation

$$n_\alpha = \frac{n_o n_e}{(n_o^2 \sin^2 \alpha + n_e^2 \cos^2 \alpha)^{1/2}} \quad (1)$$

The perpendicular component of polarization has the value n_o for all values of α .

The liquid crystal in the droplet is not perfectly aligned, but the director is configured in a manner which depends on the type of molecular anchoring at the droplet wall and the strength and direction of an applied field. Figure 1 illustrates a droplet with parallel wall alignment in the presence of an electric field (E) computed for the case where the deformation constants are equal.⁹ We may regard the overall droplet as having average indices \bar{n}_e and \bar{n}_o which depend on the director configuration in the droplet. It is clear that in the case where $n_e > n_o$, we have $\bar{n}_o > n_o$ and $\bar{n}_e < n_e$ because on the average, each component contains small admixtures of the other. For a droplet in a PDLC film, we therefore write an effective index given by

$$n_d = \frac{\bar{n}_o \bar{n}_e}{(\bar{n}_o^2 \sin^2 \theta + \bar{n}_e^2 \cos^2 \theta)^{1/2}} \quad (2)$$

where the values of \bar{n}_o and \bar{n}_e are not precisely known but depend on the director configuration in the droplet.

For droplets smaller than the wavelength of light, we can approximate the total scattering cross section⁸ σ of the droplet by $\sigma \sim (n_p - n_d)^2$.

IV. ELECTRO-OPTIC MEASUREMENTS

The optical properties and electro-optic response of PDLC light valves were measured using the simple experimental setup shown in Fig. 2. A linearly polarized, collimated beam from a helium neon laser is passed through the sample. The sample is held on a stand that may be rotated to vary the incident angle of the laser beam. The light passes through the sample to a photodiode. The output of the photodiode is linear with light intensity over the range used. An electric field of known intensity and duration is provided by application of a voltage from a pulse generator and amplifier to transparent conducting electrodes. The voltage pulse and optical response are monitored on a Tektronix 5116 storage oscilloscope and the results plotted on a HP7004B X-Y recorder.

Application of an electric field normal to the film sur-

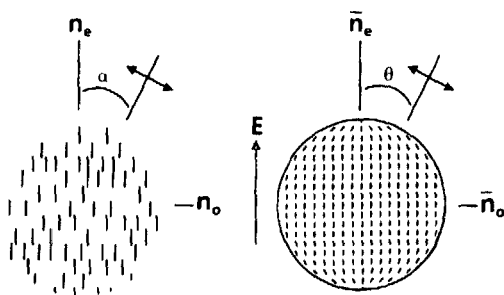


FIG. 1. Illustration of an aligned nematic and a nematic inside a droplet with parallel wall alignment in the presence of an electric field, where n_e and n_o are the refractive indices parallel and perpendicular to the direction of molecular alignment and \bar{n}_e and \bar{n}_o are those for the droplet. The direction of incident polarized light is indicated by the angles α and θ .

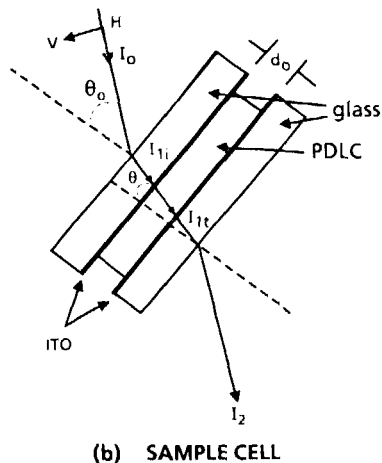
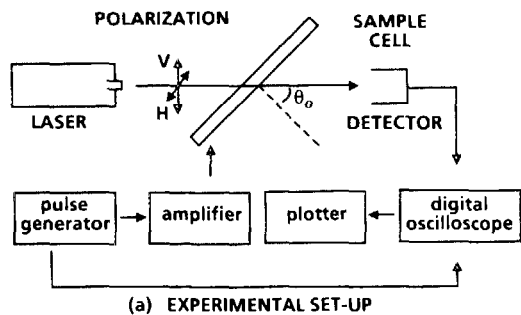


FIG. 2. (a) Experimental arrangement and (b) sample cell indicating the path of light polarized along directions V and H .

face serves to align the droplet directors parallel to the field. The transparency of the film is dependent on the angle and polarization of the incident beam and is monitored as the intensity of the transmitted beam I_2 . As indicated in Fig. 2, the polarizations H and V are those perpendicular and parallel to the plane of incidence, respectively. The direction of H is also perpendicular to the directors in the aligned droplet but parallel to the axis of sample rotation.

The angular dependence of the transmitted light intensity is governed by several factors which include (1) the Fresnel transmission coefficients¹⁰ T_1 and T_2 from the upper and lower glass-air interfaces of the sample cell, (2) the angular dependence of n_d in the case of V polarization, and (3) the increased path length ($d - d_0$) that light must travel when passing through the PDLC film at an angle θ where $d = d_0/\cos \theta$.

The effect of Fresnel reflections can be readily accounted for by the expression¹⁰

$$\frac{I_{1t}}{I_{1i}} = \left(\frac{1}{T_1 T_2} \right) \frac{I_2}{I_0}, \quad (3)$$

where I_{1t}/I_{1i} is the ratio of the transmitted to incident light intensity of the PDLC film and I_2/I_0 is the ratio of incident to transmitted light intensity of the cell. The value of I_2 is measured and I_0 held constant.

In the case of H polarization, the index variation with angle is expected to be small as the nematic director is nearly everywhere perpendicular to the direction of H . In this case, the effect of increased path length is observed. The collection angle in the experimental setup of Fig. 2 is $\sim 1.0^\circ$. If we

assume that all scattered photons do not enter the detector then simple considerations yield the equation

$$I_{1t}/I_{1i} = e^{-\beta(d-d_0)}, \quad (4)$$

where the scattering coefficient $\beta = \alpha\sigma$; α is the droplet density, and σ is the scattering cross section of a nematic droplet. In the case of V polarization, I_{1t}/I_{1i} is dominated by the index variation of n_d .

V. EXPERIMENTAL RESULTS AND DISCUSSION

A. Angular discrimination

A dc pulse is applied to 25- μm -thick PDLC samples of composition described in Table I. An electric field greater than 4 V/ μm was applied in each case; the field was of sufficient strength and duration to align the nematic droplets. In the presence of the field, the droplets became aligned and the transmitted intensity (I_2) was measured. Figure 3 shows the observed intensity as a function of angle θ_0 for both the V and H polarizations for each of the four sample cells with different values of n_p . In each case the value of I_2 is normalized to a value of 1.0 at maximum transmission intensity. It is not meaningful to compare the magnitude of I_2/I_0 among each cell as there are variations in droplet size, density, shape, and director configuration between each sample. Such features will be discussed later in subsequent work.

The angular dependence of these films for V polarized light is clearly dependent on the ratio n_p/n_o . For $n_p/n_o \leq 1$ the maximum transmission occurs at normal incidence. It is clear that the angular breadth of the transmission peak is broadest in the matched case ($n_p/n_o = 1$). This is a useful result in display design where broad viewing angles are desired. The effect of index matching on display contrast is discussed in a later section.

For the case where $n_p/n_o > 1$, the angle of maximum transmission for V polarized light is shifted to angles away from the normal. Figures 3(c) and 3(d) show the effects of two different values of n_p/n_o and demonstrate that these films can be used as angular discriminators. The angle of maximum transmission can be calculated as will be shown.

Figure 4 shows the optical response to a dc pulse of two PDLC films, $n_p \approx n_o$ and $n_p > n_o$, for different orientations of the sample. Of particular interest is the case where $n_p > n_o$. Light incident normal to the film surface, $\theta = 0^\circ$, shows a rapid turn-on time briefly passing through a maximum. The transmission through the film decreases slightly during the voltage pulse. Upon removal of the voltage, the transmission initially increases followed by a slow decay to the scattering OFF state. Such a response was reported earlier¹ but left unexplained. We show here that such a response will result under the condition $n_p > n_o$. Initial application of the voltage rotates the droplets through an orientation where $n_d = \bar{n}_p$. Because $n_p > \bar{n}_o$ the droplets pass through a maximum transmission state followed by a decrease as the value of n_d falls below n_p . The reverse occurs upon removal of the electric field. Relaxation of the droplet alignment will initially lead to an increase in n_d to a value above \bar{n}_o . The droplets will pass through the index-matched condition state of maximum transmission and then decrease to the final scattering state. At an incident angle of 40° to the normal, the

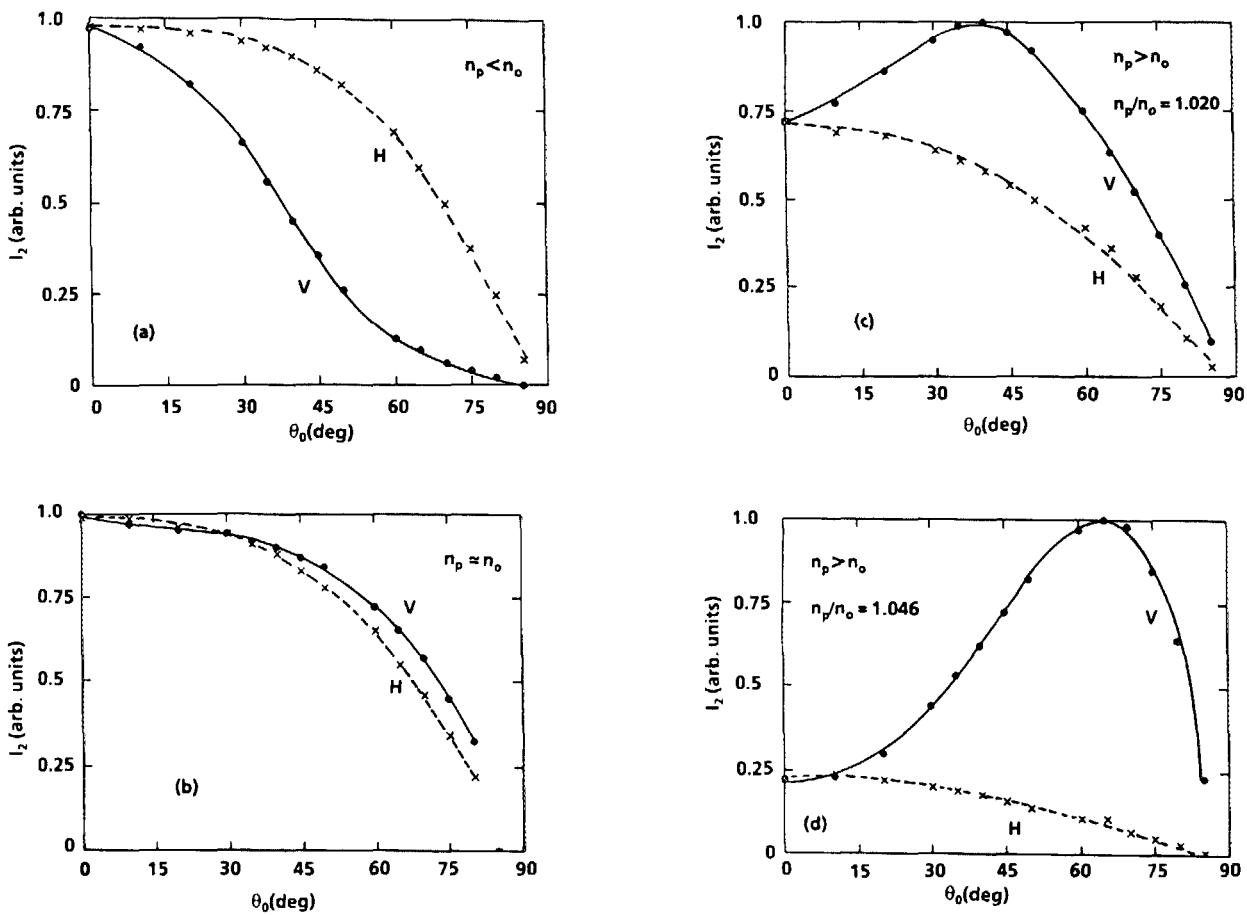


FIG. 3. Angular dependence of the transmitted intensity of V and H polarizations through PDLC films in the field ON state for four samples: (a) Epon 812 with $n_p/n_o = 0.993$; (b) polyvinylformal with $n_p/n_o = 1.006$; (c) Epon 828 with $n_p/n_o = 1.020$, and polycarbonate with $n_p/n_o = 1.046$.

transmission through the film is more than doubled. The optical rebound in the turn ON and turn OFF is not present at this angle. The film is nearly indexed matched at this angle and rotation of the droplet director from the normal will only lead to a poorer match, and a peak at the edge of the pulse is not observed. It is also noted that for the case where $n_p \approx n_o$ no peak is observed on the leading or falling edge of the pulse as expected. It is useful to remove the effects of Fresnel reflections at the air-glass interface to examine the angular dependence of the PDLC alone for V polarization. In the configuration of Fig. 2, the angular dependence of the product $T_1 T_2$ of Eq. (3) for V polarization becomes¹⁰

$$(T_1 T_2)_V = \left(\frac{4 \sin \theta \sin \theta_0 \cos \theta_0}{\sin^2(\theta_0 + \theta) \cos^2(\theta - \theta_0)} \right)^2, \quad (5)$$

where $\theta = \sin^{-1}[(\sin \theta_0)/1.52]$. Here we have taken the refractive index of the glass to be 1.52. In Fig. 5 we show the value of I_{11}/I_1 vs θ (angular response of the PDLC film) for V polarization for $n_p/n_o = 1.020$ and 1.006. Because of refraction at the planar glass-air interface, values of θ could not be achieved above 41° . In the case of $n_p/n_o > 1$, Eq. (2) can be used to determine the angle of maximum transmission, if we take the values of $\bar{n}_e \approx n_e$ and $\bar{n}_o \approx n_o$. At maximum transmission $n_d = n_p$, where from Eq. (2)

$$\sin^2 \theta = [(n_o/n_p)^2 - 1] / [(n_o/n_e)^2 - 1]. \quad (6)$$

Values from Table I yield $\theta = 23^\circ$ in agreement with Fig. 5(a). Figure 5 also shows the actual breadth of the transmission peak for the case where $n_p \approx n_o$.

The angular dependence of H polarized light in Fig. 3 is seen to be nearly the same for each sample. As described earlier, this is because in the aligned droplets the direction of H is perpendicular to the nematic director and I_2/I_0 is affected only by the Fresnel transmission coefficients and by the increased path length encountered by the light when transverse the cell at an angle θ . Because, in practice, the directors are not perfectly aligned in the droplets, some effects of index changes might be expected to be present.

Making use of Eqs. (3) and (4) for H polarization and writing¹⁰

$$(T_1 T_2)_H = \left(\frac{4 \sin \theta \sin \theta_0 \cos \theta \cos \theta_0}{\sin^2(\theta + \theta_0)} \right)^2, \quad (7)$$

it is possible to examine the effect of increased path length and measure the scattering coefficient β in Eq. (4). As the sample is reoriented in the laser beam the path length through the PDLC material, $d = d_0/\cos \theta$, increases. Figure 6 shows a plot of $\ln(I_2/T_1 T_2)_o$ vs $1/\cos \theta$ for the Epon 828 sample in which $n_p/n_o = 1.020$. The slope of the line yields a

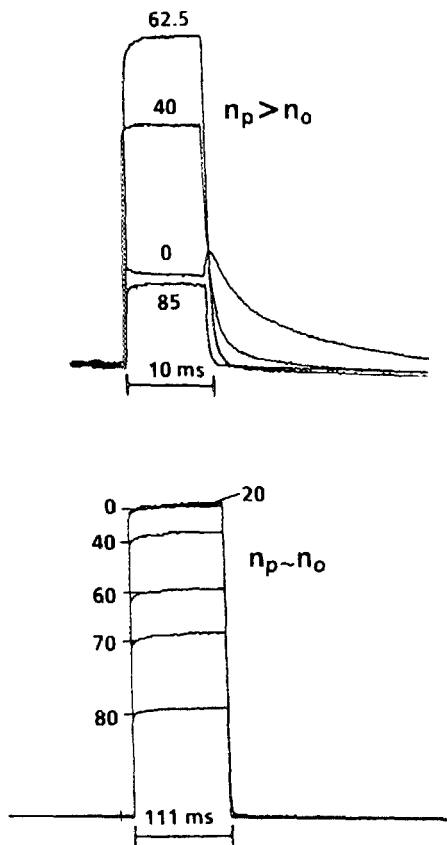


FIG. 4. Optical response of sample cells at different angles to the incident beam showing the transmitted intensity I_2 following a dc pulse of (lower) 111 ms duration for the polyvinylformal sample and (upper) 10 ms duration for the polycarbonate sample. An increase in transmission at normal incidence ($\theta_0 = 0$) following removal of the field is present in the unmatched case but not in the matched case.

value $\beta d_0 = 1.015$. The thickness of the film $d_0 = 25 \mu\text{m}$, whereby $\beta = 0.042 \mu\text{m}^{-1}$. In the Epon system, electron microscope studies show $\alpha \approx 0.5 \mu\text{m}^{-3}$ which yields a value of $\sigma \sim 0.1 \mu\text{m}^2$ for this case. This value compares favorably with that calculated for this same case (see Fig. 10 of Ref. 7) in view of the uncertainty in droplet size and applicability of the Rayleigh-Gans approximation to droplets of radius $\sim 0.5 \mu\text{m}$.

The polycarbonate sample ($n_p/n_o = 1.046$) gave a value of $\beta = 0.7 \pm 0.1$ which reflects the larger index mismatch of this sample compared to that of Epon 828. Samples of polyvinylformal and Epon 812 were so closely index matched that β could not be accurately measured but only estimated to be ~ 0.01 . Studies of β are beyond the scope of this paper and require samples well characterized for droplet size, shape, and density.

B. Voltage dependence of transmission

Figure 7 shows the light transmission for PDLC films as a function of voltage for V polarized light. In the case of the polyvinylformal ($n_p \approx n_o$) it is clear that as the voltage is increased the transparency of the film increases until a horizontal plateau is reached at about $2 \text{ V}/\mu\text{m}$. A further increase in the voltage does not change the transparency. The

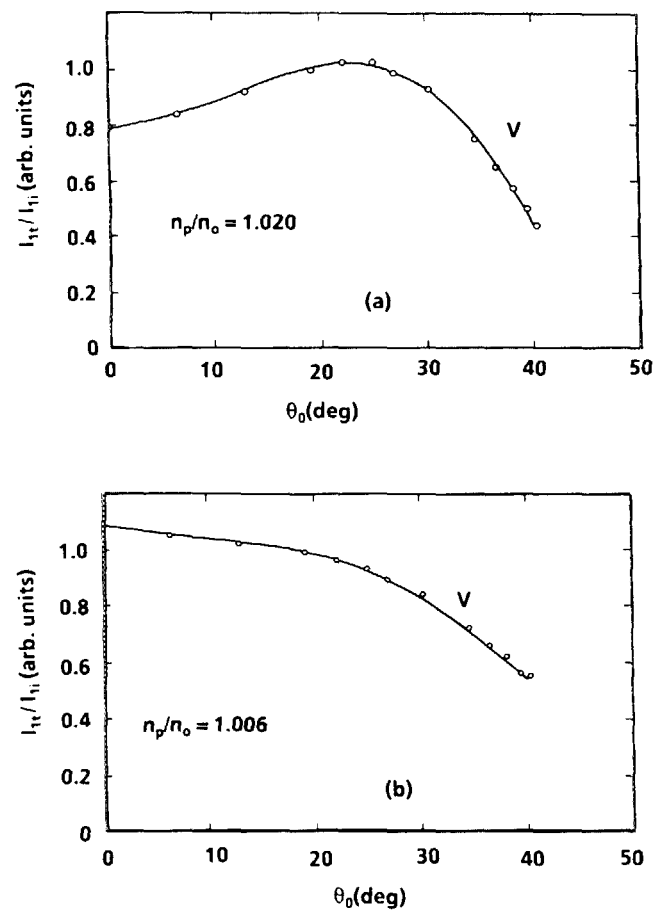


FIG. 5. Angular discrimination corrected for Fresnel reflection of PDLC films for two different values of n_p/n_o where I_t and I_i are the normalized incident and transmitted light intensities at the PDLC-glass interface as defined in Fig. 2. The samples are (a) Epon 828 and (b) polyvinylformal.

response of the Epon 828 film, $n_p > n_o$, as a function of the applied voltage shows a different character. Initially, an increase in voltage increases the transmission through the film. The transparency of the film reaches a maximum at a voltage of about $2.25 \text{ V}/\mu\text{m}$. This is the voltage at which $n_d = n_p$. As the voltage is further increased the film is less transparent

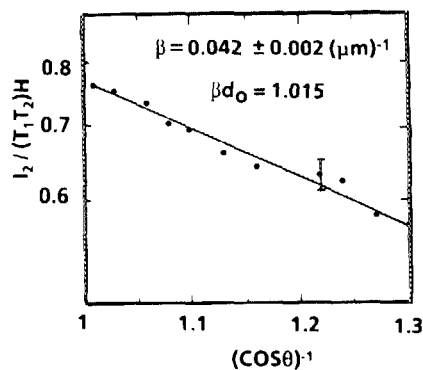


FIG. 6. Logarithmic plot of $I_2/(T_1 T_2)_H$ vs $(\cos \theta)^{-1}$ illustrating the exponential behavior of the transmitted light intensity for H polarization on the increased path length when passing through the sample at an angle. The slope of the line yields the product of the scattering coefficient β and the sample thickness d_0 , which in this case is $25 \mu\text{m}$.

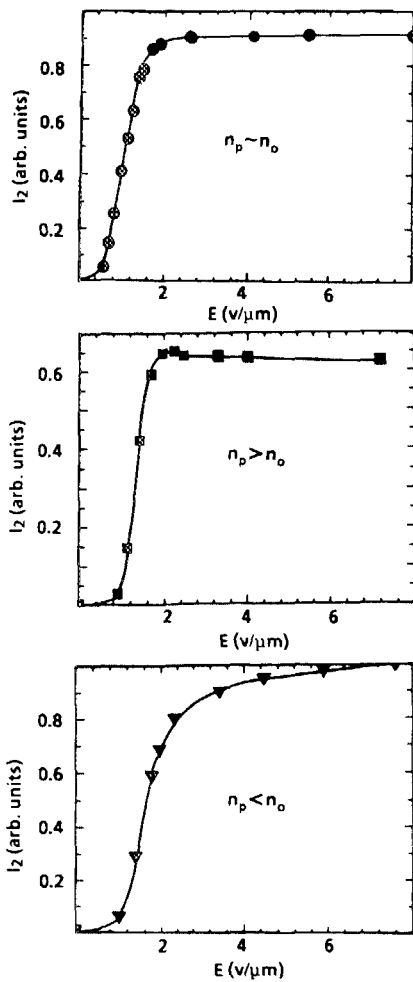


FIG. 7. Field threshold curves showing the transmitted light intensity vs applied field strength for the samples: polyvinylformal, $n_p \approx n_o$; Epon 828, $n_p > n_o$; and Epon 812, $n_p < n_o$.

because $n_d < n_p$. Such a feature is typical of threshold curves where $n_p > \bar{n}_o$. The Epon 812 film, $n_p < \bar{n}_o$, shows yet a different character. The transmission increases as the voltage is increased, never reaching a maximum. Increasing the voltage leads to n_d more closely approaching n_o but never reaching a plateau. Figure 8 shows the optical response to a

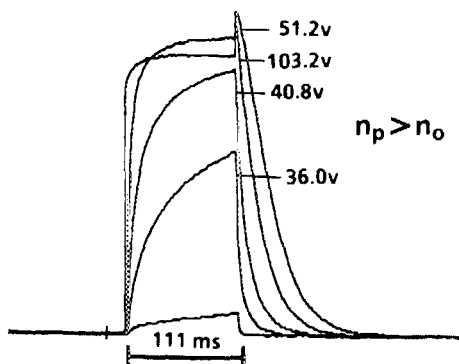


FIG. 8. Optical response of the Epon 828 sample ($n_p > n_o$) showing the transmitted light intensity during the application of a 111-ms dc pulse at voltages indicated in the figure. The sample was $25 \mu\text{m}$ thick.

dc pulse for the case of $n_p > n_o$. The effect of the increased transmission following the removal of the pulse is clearly evident. This effect only shows at the higher fields where n_d overshoots n_p again illustrating the effect in these films caused by index mismatch. It should be noted, however, that ionic charges can be present in some films which can cause a decrease in the transmission during a pulse and an increase upon removal of the pulse.

VI. CONCLUSIONS

Angular discriminating filters are shown to be possible by adjustment of the ratio n_p/n_o in PDLC films. Maximum transmission at angles away from the normal is achieved when $n_p/n_o > 1$. More sharply tuned filters appear possible by stacking films with different index matches. It is further demonstrated that maximum transmission is achieved at normal incidence when $n_p < n_o$ with the angular breadth of the transmission peak being broadest when $n_p = n_o$. These films could have application as coatings on windows where it is desired to control direct sunlight at some angles but have high visibility at others.

When the transparent state of these films is viewed under diffuse light a "residual haze" can become apparent. This haze is due to the angular dependence of the effective refractive index and is adjusted by the value of n_p relative to n_o . This feature has a strong effect on the contrast exhibited by these materials when used in information displays. Depending upon the light conditions, the optimum display contrast may not necessarily be achieved when $n_p = n_o$ but rather with a value of n_p/n_o which achieves the desired angular distribution of light transmission. A distribution in droplet size has also been reported as contributing to haze,³ but we do not believe this to be a factor in the materials reported here.

This study also provides insight into the shape of the optical response following an applied pulse. The increase in transmission following the removal of the pulse reported earlier¹ is a behavior which occurs when $n_p > n_o$. The transmission peaks as the droplet rotates through the angle where $n_d = n_p$. The peak is observed on the leading edge of the pulse but is better observed on the trailing edge perhaps because upon removal of the voltage the droplets, as a whole, rotate back to their equilibrium orientation. At the leading edge the applied field is competing with surface anchoring and the director configuration could be more scrambled.

The value of the ratio n_p/n_o is also apparent in the dependence of the transmission on applied voltage. The transmission may pass through a maximum and then decrease, reach a horizontal plateau, or continue to gradually increase with increasing voltage depending upon whether n_p/n_o is greater than one, equal to one, or less than one, respectively. A detailed study of the transmission versus applied voltage requires a better understanding of the director configuration inside the droplet.

Finally, it is shown that the scattering cross section for H polarized light is readily measured for aligned droplets by studies of the angular dependence. This could be a useful method for measuring the scattering characteristics of PDLC films. Studies of the dependence of the scattering

cross section on droplet size, shape, and direct configuration are in progress.

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