Reorientation gratings in polymer dispersed liquid crystals

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(Received 17 November 1997)

A mathematical model for the description of phase and amplitude modulation of a light beam normally impinging on a thin film of polymer dispersed liquid crystal controlled by a transversal electric field is presented. We introduce here also a simplified model to deal with the particular case of a laser beam diffracted by the superposition of sinusoidal amplitude and phase gratings. Approximations are introduced and results are discussed in an example. [S1063-651X(98)12809-X]

PACS number(s): 42.70.Df, 42.65.Hw, 61.30.Gd, 78.20.Bh

I. INTRODUCTION

In the fields of data storage and holographic materials there has recently been growing interest devoted to polymer dispersed liquid crystals (PDLCs), a dispersion of liquid crystal micrometer sized droplets in a polymeric binder [1]. Droplets appear as optically uniaxial spheres randomly oriented. When their size is close to visible light wavelength, owing to the refractive index mismatch with the surrounding medium, droplets produce a strong light scattering [2–6]. A PDLC film can be sandwiched between two ITO coated conducting glasses so that a low frequency electric field can be applied through it. Reorientation of the droplet directors causes normally impinging light to ''see'' only the ordinary refractive index of the liquid crystals; hence, if it equals the polymeric binder one, scattering also disappears for large droplet diameters. Light scattering for visible and infrared wavelengths can be avoided using small droplets [7], with diameters of the order of 100 nm. In this configuration the material behaves as an anisotropic medium with optical anisotropy controlled by the applied electric field. In a previous paper we have shown that the temperature grating, generated in a dye doped PDLC by the interference of two pump laser beams, gives rise to the superposition of an amplitude and a phase grating that can be detected by a probe beam. In this paper we study the creation of the same superposition of gratings generated in a PDLC film by the reorientation effect of a sinusoidally modulated electric field. This electric field can be due again to the interference of two laser beams. It can be the electric field of the pump beams, and in this case it can be parallel or perpendicular to the grating axis, depending on light polarization. It can also be due to the response of the polymeric matrix to the illumination gradient, and in this case it will be perpendicular to the grating axis. Both these cases are described by our model, once attention is paid to taking the liquid crystal dielectric anisotropy at the correct frequency.

II AMPLITUDE MODULATION

We consider a thin layer of PDLC between the planes $z=0$ and $z=d$ where d is the sample thickness. An electric field is transversely applied: it does not depend on z and it does not have a z component. When a light beam impinges normally on the $z=0$ plane it can be split into two components, an extraordinary one with electromagnetic field parallel to the electric one and an ordinary beam along the perpendicular direction. They undergo different scattering losses.

Assuming that the number of droplets per unit volume N_v is not high, so that we are in single scattering regime, the sample transparency for the transmitted amplitude can be expressed as

$$T=\exp(-N_v d\sigma_s/2). \tag{1}$$

$\sigma_s(E)$ is the sample scattering cross section, i.e., the droplet scattering cross section σ_d averaged over the sample length and E is the transversely applied electric field. When the impinging light polarization is parallel to the applied electric field we have an extraordinary beam. In a previous paper [8] we have shown that for $\sigma_s(E)=\sigma_s^e$ we can write

$$\begin{aligned}\sigma_s^e=&\tfrac{1}{2}\sigma_G(2kR)^2\left(\frac{\Delta n}{n_p}\right)^2\left[\frac{(n_p-n_{\rm do})^2}{\Delta n^2}-\frac{2}{3}\,\frac{n_p-n_{\rm do}}{\Delta n}+\frac{4}{15}\right]\\&+\tfrac{1}{2}\sigma_G(2kR)^2\left[-\frac{\Delta n}{21n_p^2}\,(-13n_{\rm de}-15n_{\rm do}+28n_p)S_s\right.\\&\left.+4\,\frac{\Delta n^2}{35n_p^2}\,\tilde{S}_f\right],\end{aligned} \tag{2}$$

where R is the droplet radius, $\sigma_G=\pi R^2$ is the droplet geometrical cross section, $k=2\pi/\lambda$ is the impinging beam wave number, n_p is the polymer refractive index, $\Delta n=n_{\rm de}-n_{\rm do}$, and for the droplet refractive index dependence on the droplet's order parameter S_d they hold [9]:

$$n_{\rm do}=\frac{2}{\pi}\,n_oF\left(\frac{\pi}{2},\frac{1}{n_e}\;\sqrt{\frac{2}{3}\,(n_e^2-n_o^2)(1-S_d)}\right), \tag{3}$$

$$n_{\rm de}=\frac{n_on_e}{\sqrt{\frac{2}{3}\,(n_o^2-n_e^2)S_d+\frac{1}{3}\,(n_o^2+2n_e^2)}}, \tag{4}$$

where $F(\theta,m)$ is the complete Legendre elliptic integral of the first kind and n_o and n_e are the ordinary and extraordi-

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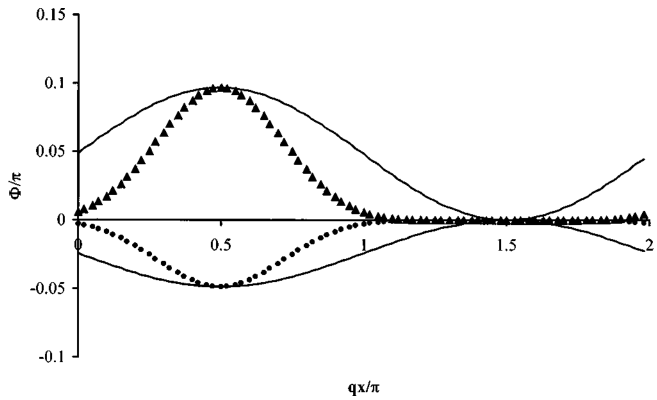

FIG. 1. Optical phase shift for a light beam normally impinging on a PDLC film undergoing modulation by a transverse sinusoidal electric field with amplitude 1 V/μm. Triangles are for the extraordinary beam, while circles are for the ordinary beam. Solid lines are the sinusoidal approximation.

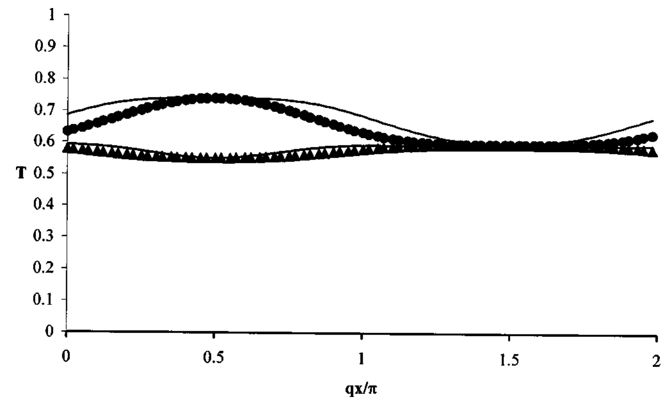

FIG. 2. Amplitude modulation of a light beam normally impinging on a PDLC film undergoing modulation by a transverse sinusoidal electric field with amplitude 1 V/μm. Triangles are for the extraordinary beam, while circles are for the ordinary beam. Solid lines are the sinusoidal approximation.

nary refractive indices of the liquid crystal, respectively. For the droplet order parameter S_d we use

$$S_d(S_s) = S_{d0}\{\exp(-C_1 S_s) + \exp[-C_2(S_s-1)]\}, \quad (5)$$

where C_1 and C_2 are sample-related parameters taking into account the dependence of the alignment of liquid crystal molecules inside each droplet on the value of S_s. C_1 takes into account the lowering of the droplet order parameter when an electric field begins the droplet reorientation, while C_2 takes into account the rise of the order droplet parameter near the saturation. If unknown, they can be put equal to zero, which is equivalent to considering a constant S_d. For the sample order parameter $\langle P_2(\cos\vartheta)\rangle_\vartheta = S_s$ we use the expression

$$S_s = \frac{1}{4} + \frac{3}{16}\,\frac{e_a^2+1}{e_a^2} + \frac{3}{32}\,\frac{(3e_a^2+1)(e_a^2+1)}{e_a^3}\,\ln\left|\frac{e_a+1}{e_a-1}\right|, \quad (6)$$

where e_a is a dimensionless reduced electric field given by [10]

$$e_a = E\sqrt{\frac{v_{\mathrm{lc}}(\epsilon_\parallel - \epsilon_\perp)}{K_d}}, \quad (7)$$

v_{lc} is the volume fraction of liquid crystal in the sample, K_d is an elastic constant per unit surface (in units of Newton per square meter) taking into account the torque, which, after the field is switched off, produces relaxation of the droplets to their original orientation, ϵ_p is the polymer dielectric permittivity, and $\epsilon_\parallel$ and $\epsilon_\perp$ are the extraordinary and ordinary liquid crystal dielectric permittivities, respectively. They depend on the applied electric field frequency and, in the visible range, are the squares of the refractive indices. Static values can usually be used to a good approximation for quasistatic frequency values, up to about 1 kHz. For $\langle P_4(\cos\vartheta)\rangle_\vartheta = \tilde{S}_f$ we use the expression [8]

$$\tilde{S}_f = \frac{7}{12} + \frac{5}{12}S_f - \frac{35}{32e_a^2}\left[\frac{2}{3} + \frac{(e_a^2-1)^2}{4e_a^2} - \frac{(e_a^2+1)^2(e_a^2-1)}{8e_a^3}\arctan\left(\frac{2e_a}{e_a^2-1}\right)\right]. \quad (8)$$

When the impinging light polarization is perpendicular to the applied electric field we have an ordinary beam, and for $\sigma_s(x) = \sigma_s^{(O)}$ we have

$$\sigma_s^O = \frac{1}{2}\,\sigma_G(2kR)^2\left(\frac{\Delta n}{n_p}\right)^2\left[\frac{(n_p-n_{\mathrm{do}})^2}{\Delta n^2} - \frac{2}{3}\,\frac{n_p-n_{\mathrm{do}}}{\Delta n} + \frac{4}{15}\right] + \frac{1}{2}\,\sigma_G(2kR)^2\left[-\frac{\Delta n}{21n_p^2}(5n_{\mathrm{de}}+9n_{\mathrm{do}}-14n_p)S_s - \frac{\Delta n^2}{35n_p^2}\tilde{S}_f\right], \quad (9)$$

where the symbols have the same meaning as before.

III. PHASE MODULATION

In a previous paper [10] for the total optical phase shift φ_θ of a light beam traveling with angle θ with the z axis we have written

$$\Phi_\theta = \frac{2\pi}{\lambda}\,\frac{dv_{\mathrm{lc}}}{\cos\theta}\,\delta n, \quad (10)$$

where λ is the wavelength, d is the sample thickness, and δn is given by

$$\delta n = n_{\mathrm{do}}\,\frac{n_{\mathrm{de}} - \sqrt{n_{\mathrm{do}}^2 + (n_{\mathrm{de}}^2 - n_{\mathrm{do}}^2)S_s\cos^2\theta}}{\sqrt{n_{\mathrm{do}}^2 + (n_{\mathrm{de}}^2 - n_{\mathrm{do}}^2)S_s\cos^2\theta}}. \quad (11)$$

Applying the same model, for the total optical phase shift Φ of a normally impinging light beam, we write

$$\Phi = kdv_{\mathrm{lc}}\delta n, \quad (12)$$

where for an extraordinary beam δn is given by

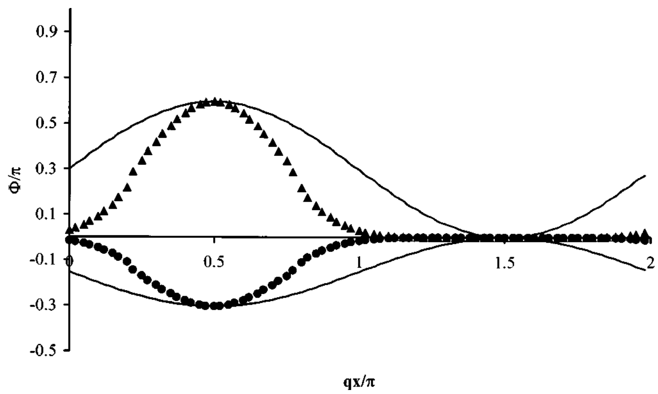

FIG. 3. Optical phase shift for a light beam normally impinging on a PDLC film undergoing modulation by a transverse sinusoidal electric field with amplitude 1.5 V/μm. Triangles are for the extraordinary beam, while circles are for the ordinary beam. Solid lines are the sinusoidal approximation.

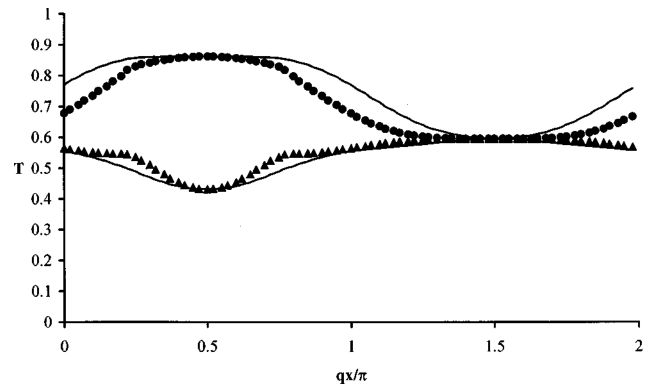

FIG. 4. Amplitude modulation of a light beam normally impinging on a PDLC film undergoing modulation by a transverse sinusoidal electric field with amplitude 1.5 V/μm. Triangles are for the extraordinary beam, while circles are for the ordinary beam. Solid lines are the sinusoidal approximation.

$$\delta n_e = \sqrt{[2n_{\rm do}^2(1-S_s)+n_{\rm de}^2(1+2S_s)]/3}, \tag{13}$$

while for a normally impinging ordinary light beam we write

$$\delta n_o = \sqrt{[n_{\rm do}^2(2+S_s)+n_{\rm de}^2(1-S_s)]/3} \tag{14}$$

IV. SINUSOIDAL GRATINGS

If a light beam impinges on a thin film of PDLC influenced by a transversal electric field that is locally variable, the complex amplitude of both its extraordinary and ordinary components can be computed by means of the model we have introduced so far. Usual Fraunhofer diffraction theory can then be used to get the far field signal. This procedure, though completely defined, can be difficult to realize. We introduce here a simplified model to deal with the particular case of a laser beam diffracted by a PDLC undergoing sinusoidal modulation by an electric field described by

$$E(x) = E_0(1+\sin qx)/2. \tag{15}$$

For our purposes the electric field can be a high frequency field of electromagnetic origin, such as the one generated by two crossed pump laser beams, as well as a static electric field, such as the one generated by sinusoidal illumination in a photoconductive polymeric matrix.

We assume the incident probe beam to be a plane wave. Its amplitude just behind the film is

$$\begin{aligned} A_t = {} & A_i U(L/2-|x|)U(L/2-|y|) \\ & \times\left(\frac{T(E_0)+T(0)}{2}+\frac{T(E_0)-T(0)}{2}\sin qx\right) \\ & \times \exp i\,\frac{\Phi(E_0)(1+\sin qx)}{2}, \end{aligned} \tag{16}$$

where A_i is the incident amplitude, and the function U defined by

$$U(x)=0 \quad \forall \quad x<0, \tag{17}$$

$$U(x)=1 \quad \forall \quad x\geqslant 0 \tag{18}$$

is used to take into account the finite size of the incoming beam. $T(E_0)$ and $\Phi(E_0)$ are given by Eqs. (1) and (12). Under this assumption the diffracted beam has a very simple analytical expression. With our sinusoidal approximation the ratio between the first order diffracted power W and the input power W_i is given by the simple analytical expression [11]

$$\begin{aligned} W/W_i = {} & \left(\frac{T(E_0)+T(0)}{2}\right)^2 J_1^2(\Phi(E_0))+\left(\frac{T(E_0)-T(0)}{4}\right)^2 \\ & \times[J_0(\Phi(E_0))-J_2(\Phi(E_0))]^2, \end{aligned} \tag{19}$$

where J_i is the first kind of Bessel function of order i.

V. DISCUSSION AND CONCLUSIONS

To have an idea of the accuracy of our approximation we consider an example. A PDLC sample of thickness $d = 10\ \mu$m is described by the following set of parameters: droplet radius $R=0.6\ \mu$m, liquid crystal refractive indices $n_o=1.511$ and $n_e=1.736$, polymer refractive index $n_p = 1.53$, glass refractive index $n_g=1.53$, $\epsilon_\parallel/\epsilon_0=18$ and $\epsilon_\perp/\epsilon_0=10$, droplets per unit volume $N_v=0.5\times 10^{18}$ m^{-3}, nematic to isotropic phase transition temperature T_{NI}

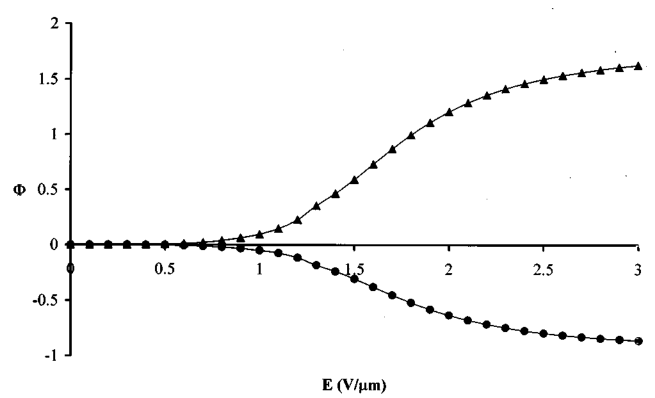

FIG. 5. Maximum optical phase shift for a light beam normally impinging on a PDLC film versus the transverse electric field value. Triangles are for the extraordinary beam, while circles are for the ordinary beam.

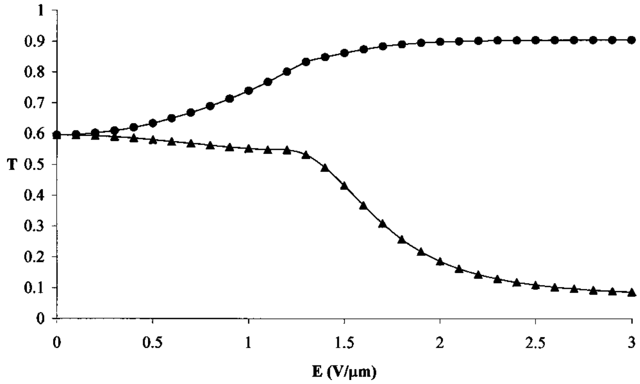

FIG. 6. Amplitude modulation of a light beam normally impinging on a PDLC film versus the transverse electric field value. Triangles are for the extraordinary beam, while circles are for the ordinary beam.

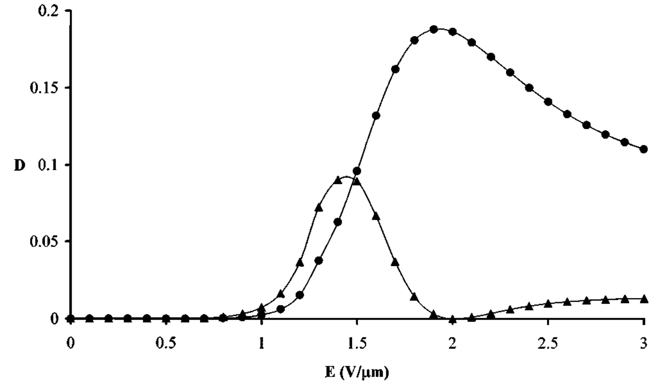

FIG. 7. Ratio between the diffracted and the incoming beam powers versus the amplitude of the transverse sinusoidal electric field. Triangles are for the extraordinary beam, while circles are for the ordinary beam.

$=54.5\,°C$, liquid crystal order parameter $S|_{T=25.5\,°\mathrm{C}}=0.611$, $K_d=44\ \mathrm{N/m^2}$, $S_{d0}=0.7$, $C_1=1$, and $C_2=14$. In Fig. 1 we show a comparison between the phase shift as computed point by point and by our sinusoidal approximation. The electric field is the one given in Eq. (15). Triangles denote the optical phase shift of an extraordinary beam, linearly polarized in the plane determined by the propagation director, in our case the *z* axis, and the direction of the transverse electric field. Circles denote the phase shift of the ordinary beam, linearly polarized in the plane determined by the propagation director and the direction perpendicular to the electric field. Solid lines are the results of our sinusoidal approximation. The maximum applied electric field is $E_0=1\ \mathrm{V}/\mu\mathrm{m}$. In Fig. 2 we see the comparison of the amplitude modulation by the same electric field. The symbol meaning, here and in the following, is the same as before. We see that the sinusoidal approximation gives a reasonably accurate description of both the transparency and phase shift. This agreement is also better for a transverse electric field $E_0=1.5\ \mathrm{V}/\mu\mathrm{m}$, as can be seen in Figs. 3 and 4. In this paper we have also given an analytical expression for $T(E_0)$ and $\Phi(E_0)$ so that no numerical simulation is required. In Fig. 5 we report $\Phi(E_0)$ versus E_0 for our sample, while in Fig. 6 we report $T(E_0)$ versus E_0. Again crosses denote the extraordinary beam and circles the ordinary one. We see a quite regular behavior with a transition between 1 and 1.5 V/μm, which is the reason why we chose these values for Figs. 1–4. A more intricate behavior is shown by the diffracted beam. In Fig. 7 we see that around the transition the extraordinary beam is higher than the ordinary one; nevertheless, a further increase in the electric field results in a strong prevalence of the ordinary diffracted beam.

In conclusion, we have introduced a mathematical model able to describe the amplitude and phase modulation of a light beam normally impinging on a PDLC film undergoing the action of a transverse electric field. Analytical expressions are given, bounding these quantities to the parameters describing the material and to the electric field. Furthermore the case of a sinusoidal modulation of the electric beam has been studied. We have shown in an example that the resulting beam modulation by the PDLC is similar to the superposition of an amplitude and a phase sinusoidal grating. Within the approximation of actual sinusoidal beam modulation by the PDLC, we have given an analytical expression for the first order diffracted beam and we have discussed it in an example.

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