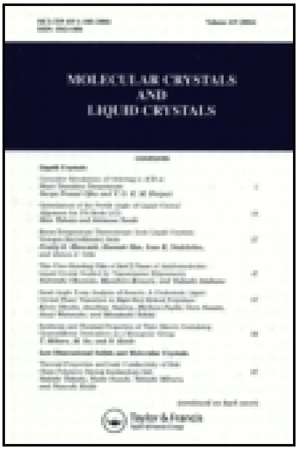


Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

Electrically Controllable Diffraction Efficiency of H-PDLC Film Composed of Ellipsoidal Liquid Crystal Droplets

V. Kubytskyi [a], V. Reshetnyak [a] & T. Galstian [b]
[a] Physics Faculty, Kyiv National Taras Shevchenko University, Kyiv, Ukraine
[b] Center for Optics, Photonics and Laser, Physics Department, Laval University, Quebec, Canada

Version of record first published: 22 Sep 2006

To cite this article: V. Kubytskyi, V. Reshetnyak & T. Galstian (2006): Electrically Controllable Diffraction Efficiency of H-PDLC Film Composed of Ellipsoidal Liquid Crystal Droplets, Molecular Crystals and Liquid Crystals, 453:1, 321-332

To link to this article: http://dx.doi.org/10.1080/15421400600656368

Mol. Cryst. Liq. Cryst., Vol. 453, pp. 321–332, 2006

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400600656368

Electrically Controllable Diffraction Efficiency of H-PDLC Film Composed of Ellipsoidal Liquid Crystal Droplets

V. Kubytskyi
V. Reshetnyak
Physics Faculty, Kyiv National Taras Shevchenko University, Kyiv, Ukraine

T. Galstian
Center for Optics, Photonics and Laser, Physics Department, Laval University, Quebec, Canada

In this work the electric field control of diffraction efficiency of holographic polymer dispersed liquid crystal (H-PDLC) film is investigated. It is supposed that H-PDLC film contains ellipsoidal bipolar liquid crystal (LC) droplets. We developed a theoretical model that combines anisotropic coupled-wave theory, Monte-Carlo simulations for director profile within droplet and statistical averaging with the orientational distribution function for droplet symmetry axes. Monte Carlo simulations are done in Lebwohl-Lasher (LL) lattice model supposing strong director tangential anchoring conditions at droplet surface. We calculated diffraction efficiency upon applied voltage for ellipsoidal droplets with different eccentricity and Gaussian orientational distribution function of droplets axes. We investigated diffraction properties of H-PDLC film for both s- and p- light polarizations.

Keywords: diffraction efficiency; holographic polymer dispersed liquid crystals; monte carlo simulation

INTRODUCTION

Much effort has been devoted recently to liquid crystal (LC) composite materials for use in volume diffraction gratings whose efficiency can be controlled by applying an electric field. The well known properties of LC, such as birefringence, optical anisotropy, the response to

Address correspondence to V. Kubytskyi, Physics Faculty, Kyiv National Taras Shevchenko University, Prospect Glushkova, 6, Kyiv 03680, Ukraine. E-mail: kubba@univ.kiev.ua

application of electrical and magnetic fields are good reasons to use it. Holographic polymer dispersed liquid crystals (H-PDLC) are intriguing materials that are usually formed by the illumination of interference patterns on a mixture of photoactive monomers with liquid crystals. After polymerization, due to local differences in photopolymerisation rates and reduced miscibility, LC domains are periodically formed (in space) with the period of the interference pattern, in areas where the light intensity was low. An electric field may then be used to manipulate the orientation of the LC so that the hologram can effectively be "hidden", making the hologram electrically switchable. Scanning electron microscopy has been used to show that the obtained grating has solid polymer regions separated by regions of high density of liquid-crystal droplets [1]. The processes of formation of switchable H-PDLC are well described in [2]. The diffraction that occurs in this material is usually in Bragg diffraction regime, which can be calculated using coupled-wave theory for thick anisotropic gratings [3]. This theory was used to describe the anisotropic diffraction properties of H-PDLC [4]. However the morphological aspects of the material (droplet form, statistics, LC configuration, etc.) were not considered in that work. Taking into account the wide field of possible applications of H-PDLC (for example in switchable focus lenses, telecommunication, optical filters, flat-panel displays), it is important to better understand those questions and to learn how to control H-PDLC by an electric field it is necessary to understand the dependence of the diffraction efficiency upon an external electric field. The present work follows this goal.

This paper is organized as follows: in Section 2 we present results of coupled wave theory for diffraction efficiency for thick H-PDLC transmission gratings. A computer simulation model is discussed in Section 3. The last section shows the systems that were simulated, explanations of obtained results, and conclusions.

DIFFRACTION EFFICIENCY OF H-PDLC FILM

In a recent paper, Montemezzani and Zgonik [3] present a two-wave coupled wave theory for volume gratings that are fabricated from birefringent materials, which is an extension to the theory of Kogelnik [5] for nonbirefringent gratings. In this work we have used this theory to calculate diffraction efficiency of transmission H-PDLC grating. Below we briefly summarize the main results of Montemezzani and Zgonik paper applicable to our system. For birefringent gratings it is assumed that the relative permittivity tensor of the grating can be written in the form

$$\hat{\varepsilon} = \hat{\varepsilon}^0 + \hat{\varepsilon}^1 \cos(Kx) \tag{1}$$

where $\hat{\varepsilon}^0$ is the average relative permittivity tensor, and $\hat{\varepsilon}^1$ relative permittivity modulation tensor, K is the grating spacing. Direct calculation of the average tensor of dielectric permittivity of the unslanted grating show that the tensor of dielectric permittivity takes a diagonal form, as in Ref. [4]. Taking into account these simplifications for H-PDLC materials, the diagonal form of the relative permittivity tensor

$$\hat{\varepsilon} = \begin{pmatrix} \varepsilon_{xx} & 0 & 0 \\ 0 & \varepsilon_{yy} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{pmatrix} \tag{2}$$

has the following components: $\varepsilon_{xx} = \varepsilon_{\parallel}$, is the dielectric permittivity for light with polarization that is parallel to the grating vector, and $\varepsilon_{yy} = \varepsilon_{\perp 1}$, $\varepsilon_{zz} = \varepsilon_{\perp 2}$ are the dielectric permittivities for perpendicular polarization. Thus in Eq. (1) we have

$$\varepsilon^0_{\perp,\parallel} = \varepsilon^{LC}_{\perp,\parallel} c + \varepsilon_{Pol}(1 - \alpha f_c), \quad \varepsilon^1_{\perp,\parallel} = \frac{2f_c}{\pi} \sin(\alpha\pi)(\varepsilon^{LC}_{\perp,\parallel} - \varepsilon_{Pol}) \tag{3}$$

where f is the volume fraction of the dispersed material (f has the form of a periodic rectangular wave that is zero in the solid polymer region and has a value f_c in the PDLC region), the width of the PDLC region is $\alpha \frac{2\pi}{K}$ (where α is a fraction $0 \le \alpha \le 1$), $\varepsilon^{LC}_{\parallel}$ and $\varepsilon^{LC}_{\perp}$ are the relative permittivities of LC for light polarized parallel and perpendicular to the director orientation, and ε_{Pol} is the relative permittivity of the polymer. To calculate values of $\varepsilon^{LC}_{\parallel}$ and $\varepsilon^{LC}_{\perp}$ we used equation for averaging dielectric permittivity over the volume of the droplet [8].

The diffraction efficiency (4) is defined to be the ratio of the power flow in the first diffracted order normal to the surface at the grating output to the power flow in the incident beam normal to the surface at the grating input, and it is given by

$$\eta = \frac{\sin^2 \sqrt{\xi^2 + \nu^2}}{1 + \xi^2/\nu^2} \tag{4}$$

with $\nu = d\sqrt{\chi_i \chi_d}$, which is a parameter that governs the coupling constant, ξ is the Bragg mismatching parameter,

$$\xi = \frac{d g_d \Delta k_i}{2\cos(\varphi_d) k_d} \tag{5}$$

and $\chi_{i,d}$ are coupling constants:

$$\chi_{i,d} = \frac{k_0 A}{4 n_{i.d}\, g_{i,d} \cos(\varphi_{i,d})} \tag{6}$$

The diffraction efficiency of an H-PDLC grating is a function of the polarization of the reading beam. In Eq. (6), $\varphi_{i,d}$ are the angles between the normal to the surface of H-PDLC and the Poynting vectors for incident and diffracted waves, $n_{i,d}$ are average refractive indexes for incident and diffracted waves, and $k_{i,d} = k_0 n_{i,d}$ are wave vectors inside H-PDLC material. The walk-off parameters $g_{\mathrm{i,d}}$ are cosines of angles between the energy propagation direction and wave vector directions in the incident and diffracted beams, while $A = e_i \hat{\varepsilon}^1 e_d = e_d \hat{\varepsilon}^1 e_i$, e_i and e_d are unit vectors along incident and diffracted electric fields, respectively. The parameter Δ is a small dephasing measure from the Bragg condition, which is defined according to Kogelnik [5]

$$\Delta = \frac{(K^2 - 2k_0 K n_i \sin(\theta_i))}{2k_0 n_i} \tag{7}$$

SIMULATION MODEL

To analyze experimental data most authors use the model of Wu, Erdmann, and Doane [11] for an isolated LC droplet with strong surface anchoring. This model considers only the mean director orientation within the droplet. We numerically simulate the director distribution inside the LC droplet to find the relative dielectric permittivity tensor of LC. The simplest model of nematic LC, which describes orientational interaction of molecules is the Lebwohl-Lasher lattice model [6], where the particles are treated as interaction sites ("spins") with continuously varying orientation but with fixed positions. There are few other models that describe properties of LC more realistically, but they are much more complicated.

In our model, the vector spins are confined to the sites of a cubic lattice. In the model system, each spin represents a closely packed group of molecules that maintain their its short range order across the nematic/isotropic phase transition. The anisotropic potential between the nearest neighbor's sites depends only on their relative orientations. The total interaction energy for our model system, consisting of nematic spins, was calculated as

$$U = -\sum_{\substack{i,j \\ i<j}} \varepsilon_{ij} P_2(\cos\beta_{ij}) - \varepsilon\xi \sum_{i=1}^{N} P_2(\cos\beta_i) \tag{8}$$

with $P_2(x) = 1/2(3x^2 - 1)$ (second order Legendre polynomial), and $\cos\beta_{ij} = u_i u_j$. Here u_i denotes the unit vector which gives the orientation of the spin located at the i-th lattice site, and β_i is the angle between the field direction and the molecular symmetry axis. The sum in Eq. (8) is only taken over the nearest neighbors. The first term in (8) is responsible for interaction between LC molecules. The ε_{ij} constant which represents the interaction strengths, is denoted by ε for nearest- neighbors

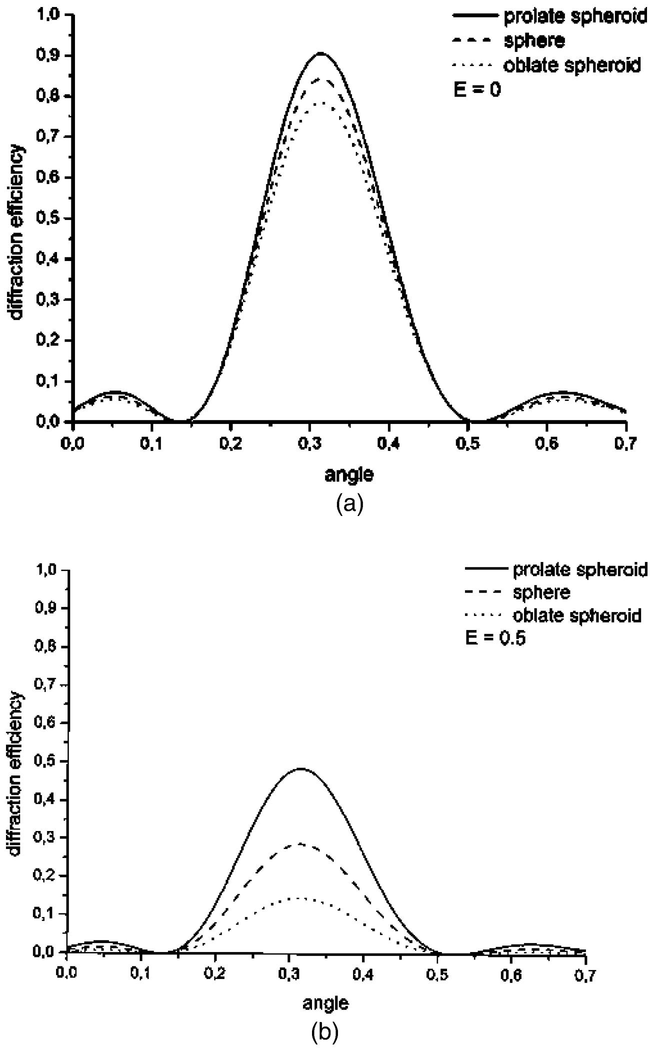


FIGURE 1 Angular selectivity of diffraction efficiency for *p*-polarized reading beam, for different electric field strength. We compare H-PDLCs composed of oblate spheroids – dot line, prolate spheroids – continuous line, and spherical droplets – dashed line.

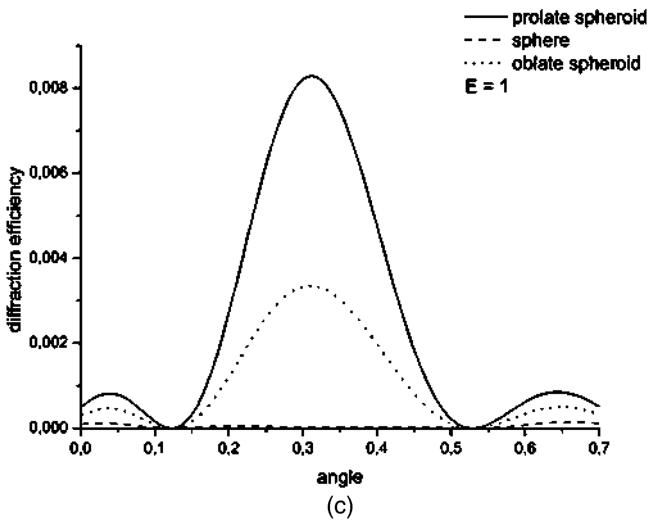


FIGURE 1 Continued.

particles i and j, and is zero otherwise. The second term in (8) describes the interaction between LC and the external electric field [7]:

$$\varepsilon\xi = \frac{\varepsilon_0}{3}\Delta\alpha \mathrm{E}^2 \tag{9}$$

The quantity ξ determines the strength of coupling with the external electric field **E**, which is assumed to be homogeneous across the drop, and $\Delta\alpha$ is the microscopic polarizability anisotropy.

RESULTS AND DISCUSSION

We constructed a model of an H-PDLC cell to study the dielectric properties of H-PDLCs containing bipolar liquid crystal droplets. The shape of the droplets can vary from oblate to prolate spheroid. In a previous work we focused on the diffraction properties of H-PDLC films composed of spherical droplets [8]. The LC director profile in each of the LC droplets is controlled by the balance of elastic energy associated with director inhomogeneity within the droplet, the anchoring energy at the droplet surface, the size and shape, and with the magnitude of the externally applied electric field. We also took into account the fact that the electric field inside an ellipsoidal droplet strongly depends on droplets shape. Calculation of the electric field in a droplet for different shapes with the same volume was done according to [9].

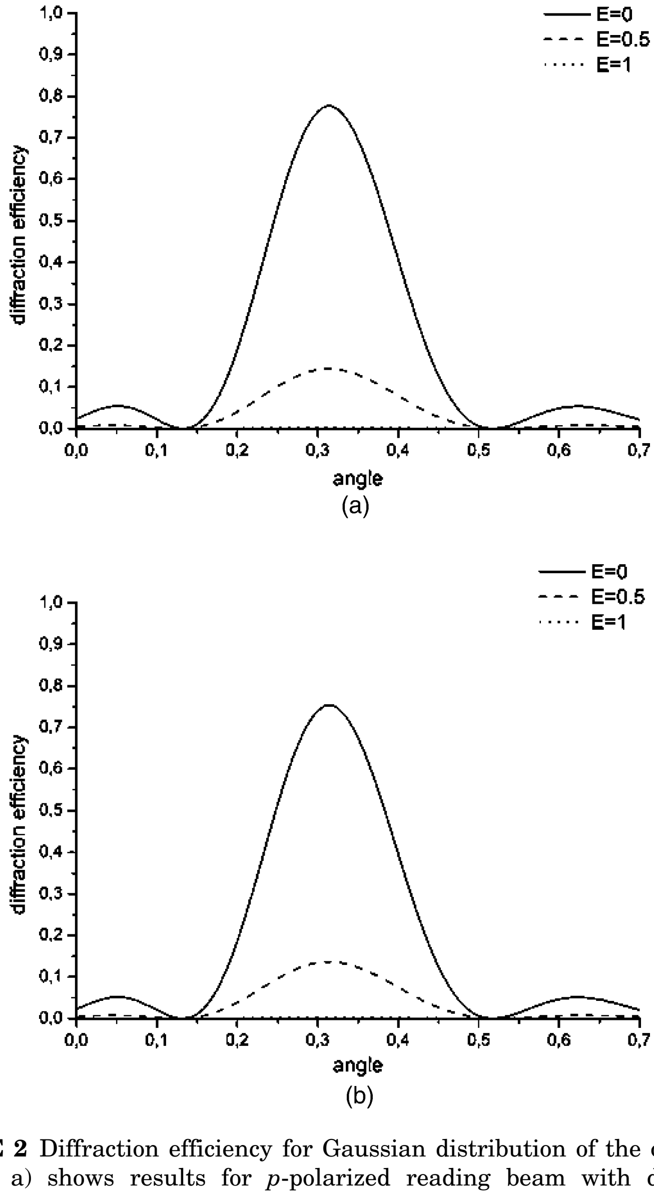


FIGURE 2 Diffraction efficiency for Gaussian distribution of the oblate LC droplets. a) shows results for p-polarized reading beam with dispersion $\sigma = 0.1$; b) $\sigma = 0.2$; c) $\sigma = 0.3$ respectively. Results for s-polarized reading beam is shown on Figure 2; d) $\sigma = 0.1$.

We calculate the relative permittivity tensor for different eccentricities of ellipsoids. For investigation of diffraction properties of the film

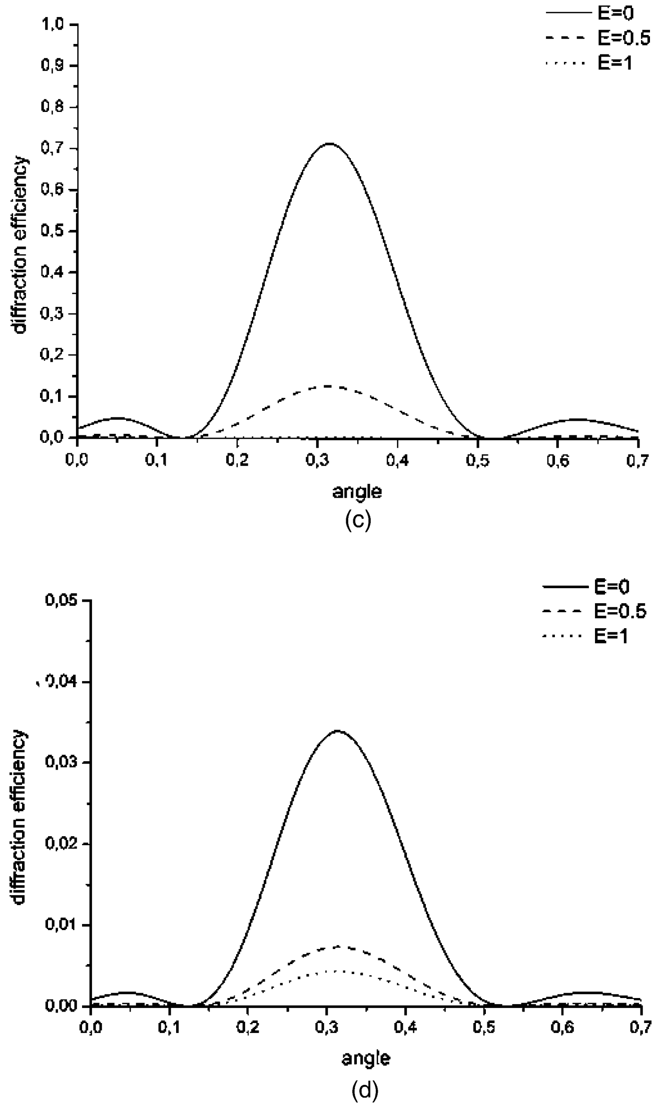
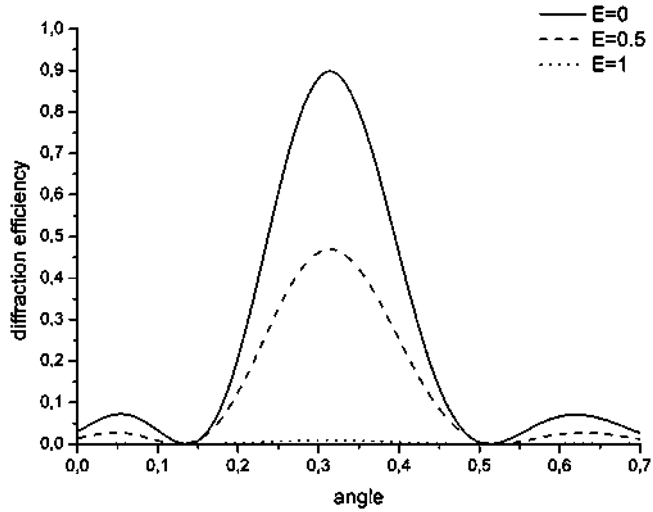
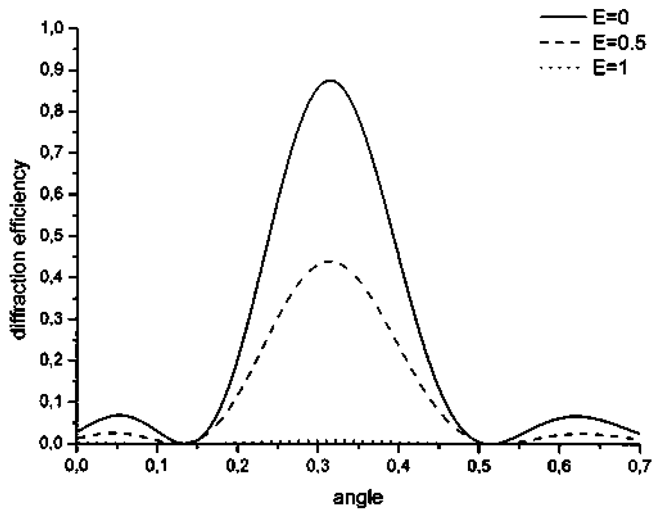


FIGURE 2 Continued.

we use the same parameters like Sutherland [10]: $\varepsilon_{\perp}^{LC} = 2.356225$, $\varepsilon_{\parallel}^{LC} = 2.8224$, $\varepsilon_{Pol} = 2.3409$, $\alpha = 0.2, f_c = 0.6$, probe light wavelength 0.633 μm, Bragg angle 0.1π, and thickness of $L = 8$ μm. Then we apply the theory of Montmezzani [3] described above, to obtain dependencies of diffraction efficiency on the angle for different values of the applied field.



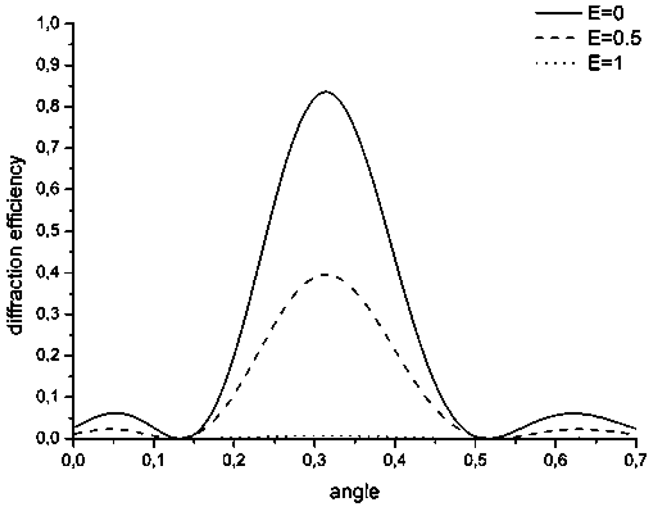
(a)



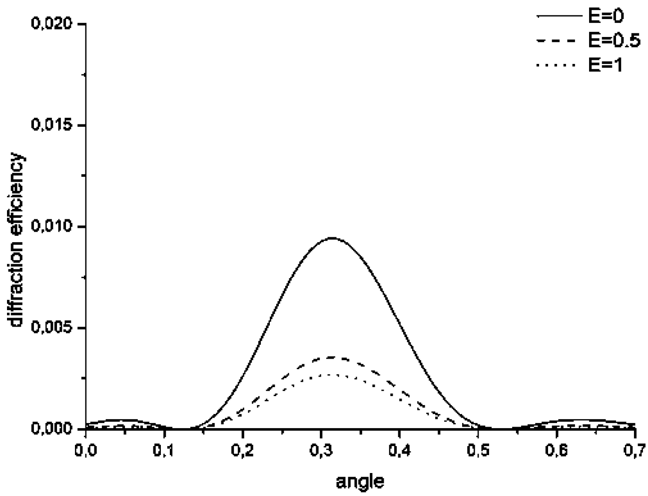
(b)

FIGURE 3 Diffraction efficiency for Gaussian distribution of the prolate LC droplets. a) shows results for p-polarized reading beam with dispersion $\sigma = 0.1$; b) $\sigma = 0.2$; c) $\sigma = 0.3$ respectively. Results for s-polarized reading beam is shown on Figure 3; d) $\sigma = 0.1$.

Figure 1 displays results for diffraction efficiency of p-polarized light on the H-PDLC film, composed of oblate spheroids (ratio of



(c)



(d)

FIGURE 3 Continued.

the semi major axes to semi minor axes equal to 4) – dot line, and prolate spheroids (ratio of the semi major axes to semi minor axes equal to 4) – continuous line, with comparison to spherical droplets – dashed line. In this case the axes of rotation of spheroids arranged along grating vector.

The same was done using a function of distribution for the axes of the rotational symmetry of LC droplets. The form of distribution function was taken from [10].

$$p(u) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{u}{2\sigma^2}\right] \tag{10}$$

We calculated the droplet dielectric tensor in "droplet" system of coordinates (axes z corresponds to axe of rotational symmetry), then we return to the laboratory system of coordinates, and calculate the average tensor elements for the droplet, using Gaussian distribution (10), where u is cosine of the angle between direction of electric field (along z in laboratory system of coordinates) and axes of rotational symmetry of the droplet.

After averaging over the orientational distribution function, we received a diagonal-form tensor for relative dielectric permittivity.

At Figure 2 we show dependencies of diffraction efficiency on external field for both p-polarized reading beam in case of the oblate spheroid LC droplets, for σ equal 0.1 Figure 2a), 0.2 Figure 2b), 0.3 Figure 2c). Diffraction efficiency for s-polarized reading beam is shown on Figure 2d), $\sigma = 0.1$. Figure 3 shows the same but for prolate spheroid LC droplets.

CONCLUSIONS

We studied dependence of diffraction efficiency of H-PDLC film composed of ellipsoidal LC droplets on externally applied electric field. Droplets of prolate and oblate spheroid forms are studied. Diffraction efficiency of the film, composed of the prolate spheroid LC droplets is higher than the diffraction efficiency of the same film, under the same voltage but composed of oblate spheroids. This difference lies in position of aligning surface of the droplet with respect to direction of an external electric field, which is almost parallel to the electric field in case of oblate droplet where molecules of LC try to align along surface, and almost perpendicular in case of prolate droplet.

Our results for Gaussian distribution of the LC droplet in the film, shows that diffraction efficiency is lower for films with higher dispersion σ of the droplets. For the films, composed of prolate spheroids LC droplets it is necessary to apply higher voltages to reorient molecules inside droplets, in order to control the diffraction efficiency.

REFERENCES

[1] Pogue, R. T., Natarajan, L. V., Tondiglia, V. P., Siwecki, S. A., Sutherland, R. L., & Bunning, T. J. (1998). *Proc. SPIE*, *3475*, 2–11.
[2] Bunning, T. J. (2000). Holographic polymer-dispersed liquid crystals. *Annu. Rev. Mater. Sci.*, *30*, 83–115.
[3] Montemezzani, G. & Zgonik, M. (1997). *Phys. Rev. E*, *55*, 1035–1047.
[4] Harbour, S., Galstian, T., Akopyan, R. S., & Galstyan, A. V. (2004). *Opt. Comm.*, *238*(4–6), 261–267.
[5] Kogelnik, H. (1969). *Bell Syst. Tech. J.*, *48*, 2909–2947.
[6] Lebwohl, P. A. & Lasher, G. (1972). *Phys. Rev. A, 6*, 426.
[7] Berggren, E., Zannoni, C., Chiccoli, C., Pasini, P., & Semeria, F. (1994). *Phys. Rev. E*, *49*, 614–622.
[8] Kubytskyi, V., Reshetnyak, V., & Galstian, T. (2005). *MCLC*, *438*, 283–290.
[9] Landau, L. D. & Lifshitz, E. M. (1975). *Electrodynamics of Continuous Media*, Pergamon Press: Oxford.
[10] Sutherland, R. L. (2002). *J. Opt. Soc. Am. B*, *19*(12), 2995–3003.
[11] Wu, B.-G., Erdmann, J. H., & Doanne, J. W. (1989). *Liq. Cryst.*, *5*, 1453.