

Polarization dependence of the nonlinear refractive index in dye-doped liquid crystals

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We report on experimental study of the polarization dependence of nonlinear refractive index in a dye-doped liquid crystal medium. Our work is based on the assumption that a polarized laser beam propagated through a nonlinear medium modifies the optical refractive index inside the material and a probe beam can experience these changes. A mathematical expression has been obtained and solved numerically for Rayleigh-Sommerfeld integral which gives the spatial distribution of the pump laser within the material. The effect of these changes on the pattern of the probe beam with different polarization has been observed.

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1. Introduction

Strong optical nonlinearity of dye-doped liquid crystals [1-7] put them among attractive substances for variety of applications such as all optical switching, modulating, and dynamic holography [8]. The origin of this nonlinearity is light induced alignment of liquid crystal molecules which is enhanced significantly by adding small amounts of dichroic organic dyes [9,10]. This effect is classified as a third order optical nonlinear effect which is based on the light induced changes in refractive index of the material. Measurement of these changes has been studied by many authors by means of z-scan technique [11,12]. This method is mainly based on far field diffraction pattern analysis. According to the previous works, when a Gaussian beam propagates through a nonlinear film, the phase distribution of the beam changes which follows by self diffraction of the incident beam. Studying the shape and the number of far field rings caused by this phenomenon can yield information about the nonlinear strength of the film [13,14]. All these works show that so far, far field self diffraction of a beam propagated through a nonlinear medium is well documented. Here, we have developed a theoretical expression for the near field diffraction pattern. In addition, we present the experimental study of the dependence of the diffraction pattern on the polarization state; these effects may be analyzed by means of a pump-probe experiment. This idea can be applicable for optical devices switched by changing the light polarization.

The diffraction theory has been introduced and explained extensively in many textbooks [15, 16, 17]. For a Gaussian and planar beam, aperture diffraction problem can be solved analytically with Fraunhofer and Fresnel approximations [18,19]. However, in the near zone where we concern, these approximations do not apply. The most general relation for a diffraction problem can be expressed

by Kirchhoff and Rayleigh-Sommerfeld integral alternatively [20]. Although the former integral is workable for many applications, it causes some problems [18, 19]. So, we opt for the latter case.

Theory

Let us consider the case in which a wave is diffracted at aperture S_0 for normal incidence. According to Rayleigh- Sommerfeld theory, the resultant wave in the near vicinity of the aperture is:

$$E(P) = \frac{1}{2\pi} \iint_{S_0} E(P_0) \cos(\vec{n}, \vec{r}) \left(\frac{1}{r} - jk \right) \frac{\exp(jkr)}{r} ds_0$$

Where $E(P_0)$ is the incident wave,

$$r = \sqrt{(x-x_0)^2 + (y-y_0)^2 + L^2}$$

$\cos(\vec{n}, \vec{r}) = L/r$, and L is a distance between aperture and observation plane.

Now, assume that the incident wave is a Gaussian-Beam. That is:

$$E(\rho_0, L) = E(0) \exp\left(-\frac{\rho_0^2}{w^2}\right) \exp\left(\frac{-jk_0 n_0 \rho_0^2}{2R}\right) \exp[-j\varphi(\rho_0, z)]$$

where ρ_0 is radial distance from axis of the beam. w and R are well-known beam parameters:

$$w = w_0 [1 + (z/z_R)^2]^{1/2} \quad R = z [1 + (z_R/z)^2]$$

Parameter $\varphi(\rho_0, z)$ is the extra phase that beam experiences when passes the nonlinear film of length dl [13]:

$$\varphi(\rho_0, z) = k_0 n_2(dl) I \exp\left(\frac{-2\rho_0^2}{w^2}\right)$$

$$E(P) = \frac{1}{2\pi} \iint_{s_0} E(0) \exp\left(-\frac{\rho_0^2}{w^2}\right) \exp\left(\frac{-jk_0 n_0 \rho_0^2}{2R}\right) \exp[-jk_0 n_2(dl) I \exp\left(\frac{-2\rho_0^2}{w^2}\right)] \cos(\vec{n}, \vec{r}) \left(\frac{1}{r} - jk\right) \frac{\exp(jkr)}{r} ds_0 \quad (1)$$

It should be noted that, we have neglected all the absorption effects (linear and nonlinear) since the cell is extremely thin. The intensity distribution is:

$$I(p) = |E(p)|^2$$

We solved the above-mentioned integral “Eq. (1)” for $L =$ cell thickness by means of Trapezoid method. Since the cell is extremely thin ($7.9 \times 10^{-3} \text{ mm}$), it is supposed that the intensity distribution inside the cell does not change significantly (mean field approximation) and equals the mean value of the incident and outcome waves right after the cell.

So, change in the nonlinear refractive index inside the substance becomes:

$$\Delta n_{mf} = n_2 I_{mf}$$

Where I_{mf} stands for mean field approximation.

Numerical results for different value of $\Delta n = n_2 I$ have been depicted in Fig. 1.

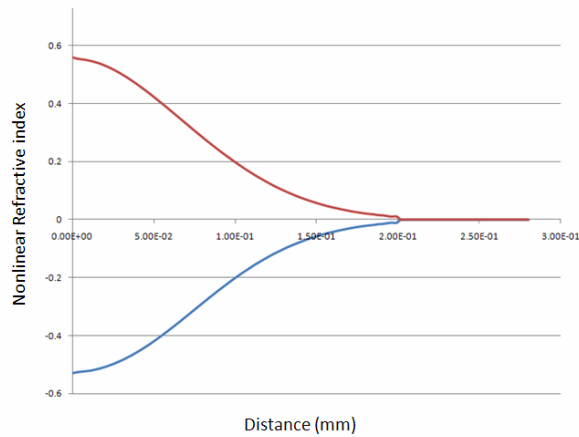


Fig. 1. Nonlinear refractive index distribution inside the material (Δn_{mf}) for different nonlinear strength (Note that the values of Δn , ± 0.54 , refer to 8 diffraction rings in the far field [13]). $\lambda = 532 \text{ nm}$, $z = 2 \text{ mm}$, $w_0 = 0.1 \text{ mm}$, $n_0 = 1.5$, $dl = 7.9 \mu\text{m}$

2. Experimental

In this experiment, nematic LC material, 40-n-pentyl-4-cyanobiphenyl (5CB, Merck), was used. The LC was

where dl is the thickness of the film.

Putting all these relation in Rayleigh- Sommerfeld integral, one obtains:

doped with Methyl Red, $C_{15}H_{15}N_3O_2$ (0.3 weight percentage), as a dye with high anisotropic response to polarized light. The reorientation of the dye molecules, as a result of their large dipole moment and cis- trans isomerisation, results in an enhancement in the reorientation of nematic LC molecules⁹.

A standard cell (thickness $7.9 \times 10^{-3} \text{ mm}$) with planar alignment was filled with LC-dye mixture by means of the capillary effect.

As shown in Fig. 2, the experiment was carried out with a single-mode (TEM00) CW He-Ne laser (633nm, power 3mw) as a probe beam and a second harmonic CW Nd:YAG laser (532nm, maximum power 100mw) as a pump beam. Note that the pump wavelength is in the region of the dye absorption. So, the Photoisomerisation process is significantly enhanced. The polarization of the pump beam is parallel to the direction of the molecules in the cell before illumination. Lenses 1 and 2 are used to focus the pump and probe beams, respectively. The probe beam is focused exactly over the centre of the pump beam spot on the sample. Polarization state of the probe beam can be altered by polarizer P . The spatial beam intensity profile distribution was observed by a CCD camera. The far-field pattern was recorded for two different cell positions, which were symmetric with respect to the lens focal plane ($z = \pm 10 \text{ cm}$). An optical filter was placed before the CCD aperture to filter out the pump wavelength.

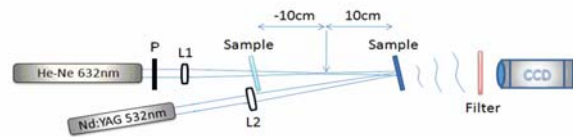


Fig. 2. Experimental setup for measurement of polarization dependence of dye doped liquid crystals nonlinear response.

3. Results and discussions

In the theoretical section, Rayleigh- Sommerfeld integral for the situation in which a Gaussian beam propagated through a nonlinear medium was calculated. We used mean field approximation to evaluate the refractive index modification inside the sample. In addition, we investigated the effect of changes in the

polarization of the probe beam, which is propagated through this medium, on the far field diffraction pattern.

As it was calculated in the theoretical section and showed in figure 1, the intensity distribution of a laser (pump laser beam) inside the thin nonlinear medium (focusing or defocusing medium) has a Gaussian profile. Because of the third order nonlinear response of the

medium, optical refractive index is modified. Consequently, the probe beam experiences an extra phase shift which has Gaussian profile too. This effect can be observed by examining of a probe beam propagated through this medium.

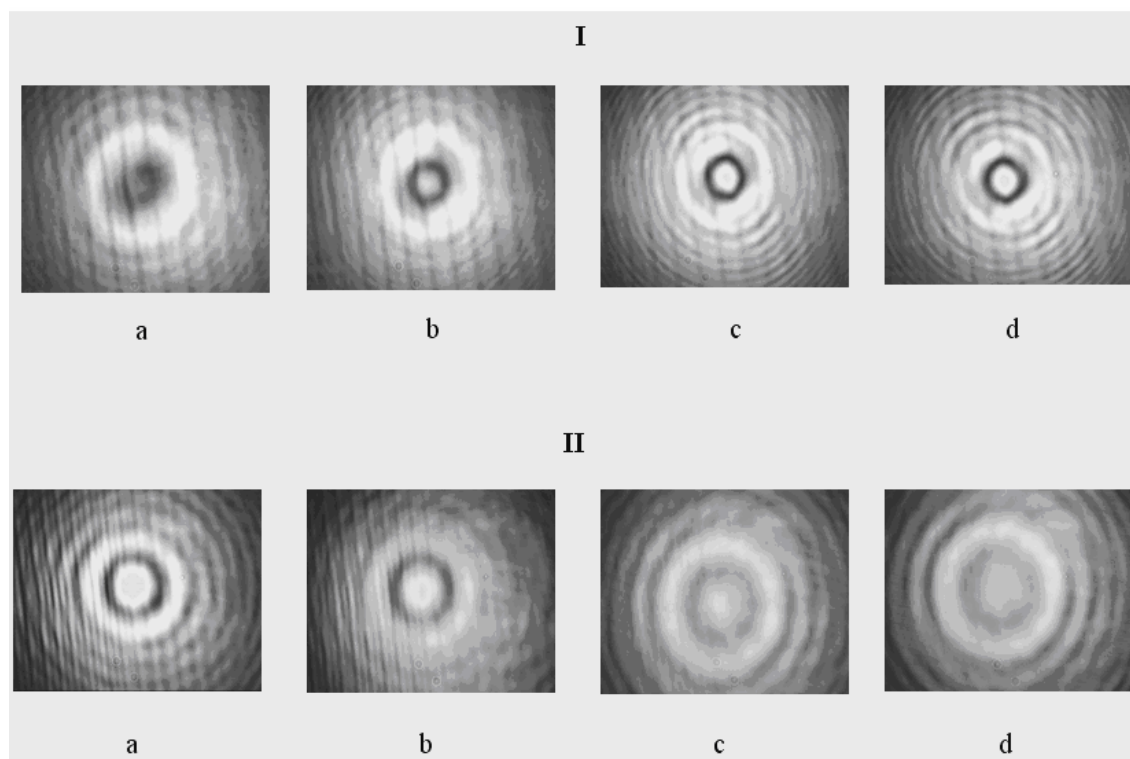


Fig. 3. Diffraction rings when the nonlinear film was placed **I**) before the beam waist **II**) after the beam waist. Letters **a,b,c,d** refer to the polarization angles 0, 30, 60, and 90 of the probe with respect to the pump beam, respectively. (The linear interference patterns, visible in some images, are due to the multiple interferences by two sides of the cell).

Fig. 3 shows the experimental results of the polarization dependence of diffraction rings in the far field. Part **I** (**II**) refers to the situation in which nonlinear medium was placed before (or after) the waist of the probe beam. Letters **a** through **d** refer to polarization difference between pump and probe beams. These results may be interpreted as following; as we have known from the previous works^{13, 21}, two different behaviour can be observed for the far-field patterns. The first case occurs for a divergent beam in self-defocusing medium ($z > 0, \Delta n < 0$) or for a convergent beam in a self-focusing material ($z < 0, \Delta n > 0$). The second behaviour is obtained for a convergent beam propagating through a self-defocusing medium ($z < 0, \Delta n < 0$) or alternatively for a divergent beam in a self-focusing medium ($z > 0, \Delta n > 0$). In the first case, brighter rings surround the central ring as in the case **a** and **b** in part **I** and **c** and **d** in part **II** of figure 3. In this case, the number of rings

increases linearly by increasing the nonlinear refractive index.

In the Second case, central ring is brighter and weaker rings surround it as in the case **c** and **d** in part **I** and **a**, **b** in part **II** of figure 3. The origin of these behaviour can be found in rotation of liquid crystal molecules induced by polarized light; the polarized pump beam causes that liquid crystals rotates perpendicular to the direction of the polarization (Photo isomerisation). So, the probe beam whose polarization is near the pump polarization (30 and 60), sees the ordinary refractive indices which is obviously less than the extraordinary refractive indices of the molecules. Consequently, the change in the optical refractive index will be negative. A similar elucidation can be used to justify the positive nonlinear refractive index seen by the probe whose polarization state is about perpendicular to the pump beam (60 and 90).

4. Conclusion

We calculated the distribution of light and nonlinear refractive index inside dye-doped liquid crystal by solving the Rayleigh-Sommerfeld integral. Besides, we evaluated optical polarization dependent response of this material by studying the far field diffraction pattern. The results may be helpful for all optical switches that work by changing of light polarization state.

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