Opto-optical modulation in *N*-(*p*-methoxybenzylidene)-*p*-butylaniline

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A method of opto-optical modulation in liquid crystals is reported. An Ar^+-laser beam is employed to modulate a second He–Ne laser. The highest frequency achieved was 1.5×10^3 pulses per second with input modulating powers smaller than 10 mW. A homeotropic *N*-(*p*-methoxybenzylidene)-*p*-butylaniline liquid-crystal cell was employed as the nonlinear medium.

It is a well-known fact that optical-field-induced refractive-index changes have an appreciable effect on laser-beam propagation in a nonlinear medium. Effects such as self-focusing and self-phase modulation have been the object of several papers treating nematic liquid crystals as nonlinear materials.[1–9] Studies have shown that the intensity-dependent refractive index associated with optically induced molecular reorientation in a typical nematic crystal, such as *N*(*p*-methoxybenzylidene)-*p*-butylaniline (MBBA), is extremely large. Third-order nonlinear optical processes, such as degenerate four-wave mixing and wave-front conjugation, are easily observable with lasers of moderate power (intensity of the order of 10 W/cm^2).[4,10] Two groups have recently published their observation of optical-field-induced effects in the nematic phase of liquid crystals. Because of the large nonlinearity, self-focusing of the laser beam in these experiments was observed.[1] In every case, the beam characteristics change drastically when the beams pass through even a thin sample of such materials. Some research has been done in this area. Shen *et al.*[6] have reported the observation of a multiple-ring pattern of laser diffraction from a nematic-liquid-crystal film. The phenomenon is shown to be the result of spatial self-phase modulation that is due to the laser-induced Freedericksz transition. Moreover, it has been shown that the dynamic behavior of this optical-field-induced transition is also analogous to that of the dc case.[5] The initial response of the induced molecular reorientation to the laser switch-on and the longer time response to the laser switch-off are both exponential with relaxation times τ_{on} and τ_{off}. The values given by Shen *et al.*, working at high Ar^+-laser intensity (350 W/cm^2), are close to 100 sec, showing good agreement with their theory. They used a He–Ne laser to probe the optical birefringence resulting from the induced molecular reorientation.

In this Letter we extend some experimental results reported previously by us[11] concerning qualitative and quantitative measurements of the opto-optical modulation of a laser beam by another laser employed as a modulator. A thin film of MBBA was employed as a nonlinear material.

The experiment uses a cw argon-ion laser focused to an e^{-2} diameter of around 164 μm at the homeotropically aligned liquid-crystal cell, as depicted in Fig. 1. The optical propagation is at an angle of 0° with the liquid-crystal molecule axis. A second, He–Ne laser forms an angle of about 40° with respect to the Ar^+ laser. Another lens was used to position this second laser beam while controlling its spot size on the sample. The relative beam-diameter sizes at the liquid-crystal cell are given by

$$w/a \simeq (\lambda_d f_1 w_f)/(\lambda_f f_2 w_d) + m(\pi w_d w_f)/(\lambda_s f_1 f_2), \qquad (1)$$

where f_1 and f_2 are the focal lengths of the lenses focusing the He–Ne and Ar^+ lasers, respectively, and m is the distance between the focus position for lens f_1 and the middle of the liquid-crystal cell. λ_d and λ_f are the He–Ne and Ar wavelengths, respectively. w_f and w_d are the Ar- and He–Ne-laser-beam radii, respectively. In our experimental setup, Eq. (1) is therefore

$$w/a \simeq 0.4 + 0.244m. \qquad (2)$$

Both lasers, 5145-Å Ar^+ and 6328-Å He–Ne, are linearly polarized with the same polarization direction, which is orthogonal to the plane containing both beams. The maximum output power for the Ar^+ and He–Ne lasers was 25 and 1 mW, respectively.

The samples studied have a separation between plates smaller than 120 μm. The homeotropic alignment was obtained by HTAB surface treatment. The experiments were carried out at room temperature (about 25°C), i.e., within the nematic range for MBBA.

We projected both laser beams onto the same screen,

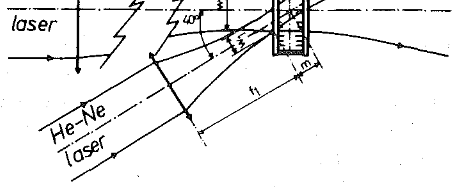

Fig. 1. Experimental setup. LCC, homeotropic liquid-crystal cell. Other symbols are defined in the text. f_1 = 5 cm, f_2 = 25 cm.

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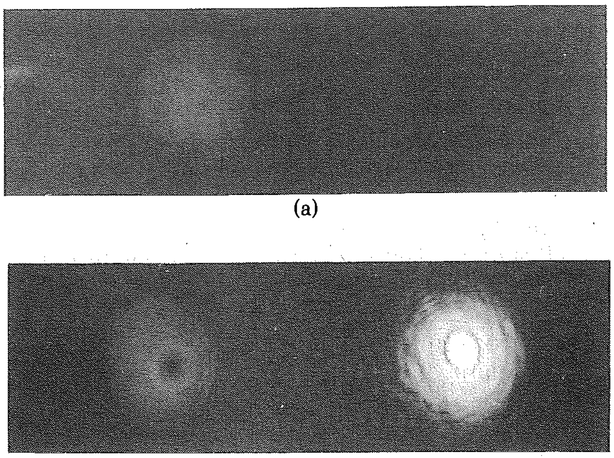

(a)

(b)

Fig. 2. Experimental results. Diffracted laser beams obtained: (a) He–Ne without Ar^+, (b) He–Ne (left) with Ar^+ (right). Ar^+-laser power, 10 mW. He–Ne-laser power, 1 mW.

which was situated about 1 m from the nematic cell. A photodetector was placed at the middle of the He–Ne image to register the intensity variations.

When no Ar^+ laser was applied to the liquid-crystal cell, the projected He–Ne laser beam had the usual Gaussian intensity profile, as could be observed by a transverse displacement of the photodetector. The same situation was obtained when the Ar^+ laser was turned on, as long as power densities were smaller than 6.3 W/cm^2, corresponding to focused intensities at the cell smaller than 60 W/cm^2. Larger powers for the Ar^+ laser gave rise to self-focusing, as had been shown by Khoo.[1] The divergence of the Ar^+ beam after crossing the sample progressively decreases as the intensity is increased, and concentric rings begin to appear. Recent studies have shown that these rings are due to the large phase modulation of the laser owing to the birefringence of the tilted molecules. The appearance of these rings signals that a large molecular reorientation has occurred.

If the focused He–Ne laser now crosses the self-focused zone, its focus being at a distance between 5 mm and 3 cm from the middle of the liquid-crystal cell, a different intensity profile is obtained at the screen. Ar^+-laser intensities slightly larger than 70 W/cm^2 reduce the He–Ne-laser-beam intensity at the center of its projection onto the screen, causing the appearance of an small bright ring around a dark center. Figure 2 is a picture of such a situation. As one can see, the initial Gaussian profile no longer appears. Use of larger Ar^+-laser intensities intensifies this effect. A black dot appears at the center, as does a clear ring around the center. When the Ar^+ intensity reaches a value of about 130 W/cm^2, rings such as the ones reported by Shen appear in the Ar^+ pattern. A similar effect occurs with the He–Ne laser but with a much smaller contrast between maxima and minima. The same black dot remains at the center of the projected image. The energy has been transfered from the initial Gaussian profile to the surrounding ring.

An important point that needs to be considered is the relative size at the cell interaction zone of the two propagating laser beams. The phenomenon described above, i.e., a black spot at the center of the emerging He–Ne beam, appears at a given diameter ratio between the Ar^+ and the He–Ne beams. Other ratios give a different diffraction image. This result can be described by a model similar to the one employed previously[6] to describe the laser-induced diffraction rings from a nematic-liquid-crystal film. We can describe both laser beams by the following equations:

$$E_f(\rho, 0) = E_0 \exp(-\rho^2/a^2), \tag{3}$$

$$E_d(\rho, 0) = E_1 \exp(-\rho^2/w^2), \tag{4}$$

where a and w are the Ar^+- and He–Ne-laser waist radii, respectively, as before. Above a certain threshold, the Ar^+-laser field E_f will reorient the direction of the molecular alignment through the medium. This leads to a local refractive-index change $\Delta n(\rho)$ in the medium seen by both lasers and to two corresponding phase shifts. The new refractive index will be given by

$$n(\rho) = n_0 + n_2 E_0{}^2 \exp(-2\rho^2/a^2). \tag{5}$$

The outgoing field for the He–Ne-laser beam after it crosses the liquid-crystal cell will be given by

$$2E_d(\rho) = E_0 \exp(-\rho^2/w^2) \exp i[\psi_0 \times \exp(-2\rho^2/a^2)] + \text{c.c.} \tag{6}$$

For a large cell-to-screen distance R we have

$$2dE_p = E_d(\rho)/R \exp[i(\omega t - kR)] dS + \text{c.c.} \tag{7}$$

Hence the total field will be

$$2E_p(q) = 2\pi E_0 \int_0^\infty \rho J_0(kq\rho/R) \exp(-\rho^2/w^2) \times \exp\{i[\psi_0 \exp(-2\rho^2/a^2)]\} d\rho + \text{c.c.}, \tag{8}$$

where q is the radial distance from the center at the projected diffraction image, R is the distance between the liquid-crystal cell and the screen, and

$$\psi_0 = \frac{2\pi d}{\lambda} n_2 E_0{}^2. \tag{9}$$

After normalizing with $u = \rho/w$ and $r = 2\pi wq/\lambda R$, we obtain

$$2E_p(r, \psi_0) = 2\pi w^2 E_0 \int_0^\infty u J_0(ru) \exp(-u^2) \times \exp(i\{\psi_0 \exp[-(w/a)^2 2u^2]\}) du + \text{c.c.} \tag{10}$$

This expression can be converted to

$$2E_p(r, \psi_0) = \pi w^2 E_0 F(r, \psi_0), \tag{11}$$

with

$$t = \exp(-u^2) \tag{12}$$

and

$$F(r, \psi_0) = \int_0^1 J_0[r \ln^{1/2}(1/t)] \times \exp[i\psi_0 t^2 (w/a)^2] dt + \text{c.c.} \tag{13}$$

These values are, in our case, $d \simeq 100\ \mu$m, $\lambda = 6328$ Å,

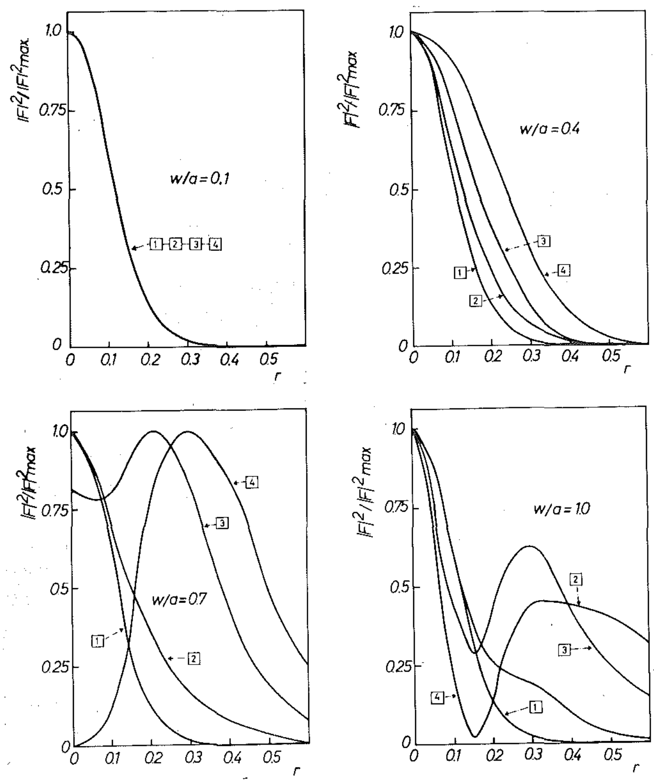

Fig. 3. Calculated diffracted wave fields for the He–Ne laser, different intensity values for the modulating Ar^+ laser, and different relative spot diameters. ψ_0 and r are defined in the text. $\psi_0 = 1$, 0 rad; 2, 1π rad; 3, 1.5π rad; 4, 2π rad.

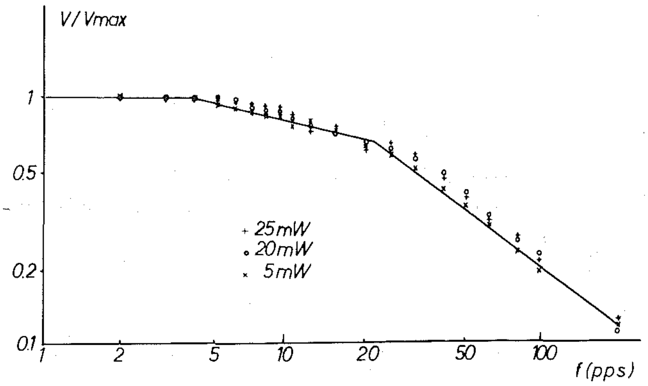

Fig. 4. Relative electric response at the diffracted He–Ne-laser-beam center as a function of the chopper frequency for three different Ar^+-laser powers.

$n_2 \simeq 10^{-3}$ cm^2/W, and $R \simeq 1$ m. We have considered cylindrical symmetry.

This diffraction pattern is pictured in Fig. 3 for w/a = 0.1, 0.4, 0.7, 1.0 and for different values of ψ_0. In each case represented, the maximum value has an assigned value of unity.

A further point needs to be considered, namely, that concerning thermally induced refractive-index changes. This effect has been studied by Khoo.[4] Its influence, as has been shown, can alter the threshold intensity and the dynamic behavior. Some of the anomalies that we have observed, and which are described below, can be due to this effect.

To observe the dynamic behavior of this phenomenon, a chopper was placed at the Ar^+-laser beam before the beam crossed the liquid-crystal cell. A photoelectric detector was situated at the center of the diffracted He–Ne-laser beam. The results are shown in Fig. 4, in which the relative electric signal obtained as a function of the chopper frequency has been represented. Three different Ar^+-laser intensities were used, namely, 6, 24, and 30 W/cm^2. Similar figures have been obtained in each case. Three main regions appear. They are characterized by three different slopes. The first region, up to frequencies of about 4 pulses per second (pps), is mainly horizontal. The second one has a slope of 3.7 dB per decade and reaches 20 pps. The third region keeps a slope of 6.6 dB per decade up to the highest frequency obtained, near 1.5×10^3 pps. In Fig. 4 the highest frequency shown is 200 pps because higher values give a relative response smaller than 0.1.

The existence of these three regions implies three different kinds of behavior whose nature deserves further study. The slope of every region has to be related to the response time of the phenomenon. Our observations are that several phenomena are taking place at the same time. In fact, a rectangular modulation of the Ar^+ beam gives rise to a modulated He–Ne-laser-beam output following no obvious exponential law.

In conclusion, we have shown that opto-optical modulation of a laser beam is possible by using nematic liquid crystals as nonlinear media. This phenomenon should be useful in applications that require a simple laser device with moderate peak-power outputs. Modulated frequencies higher that 1.5×10^3 pps can be obtained. This value is in good agreement with previous experiments with liquid-crystal modulating devices if the cell thickness is taken into account.[12]

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