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Self–focusing effects in a nematic liquid crystal at 10.6 μm

S. Brugioni[a] and R. Meucci

Istituto Nazionale di Ottica Applicata, Largo E. Fermi 6, 50125 Florence, Italy

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Abstract. E7 is a room temperature nematic liquid crystal that presents both a high temperature nematic–to–isotropic transition and a high positive dielectric anisotropy. The optical characteristics of this liquid crystal have been widely investigated in the visible region of the spectrum, while less experiments have been performed in the middle infrared spectral region, where, nevertheless, potential applications are important. We investigate the self–focusing of a middle infrared laser beam when it passes through a film of E7. We also take into account the heating of the liquid crystal film caused by the partial absorption of the laser light by means of a thermographic technique. A theoretical interpretation of the self–focusing phenomenon is given in terms of the reorientation of the molecules with respect to their unperturbed direction and an estimation of the average value of the elastic constants of the liquid crystal is given by fitting the experimental data with the theoretical model.

PACS. 42.70.Df Liquid crystals – 42.65.-k Nonlinear optics – 42.55.Lt Gas lasers including excimer and metal-vapor lasers

1 Introduction

One of the characteristic phenomena of nonlinear optics is the self–focusing of a laser beam that traverses a medium whose refractive index depends on the laser light intensity [1–3]. Consider for example a Gaussian shaped laser beam impinging on such a nonlinear medium. As a first approximation the refractive index can be assumed to be $n = n_0 + n_2 I$ where I is the laser light intensity and where n_2 is of the order of 10^{-3} cm^2/W [4]. In the central part of the spot the light intensity is larger than in the outer one. As a consequence the refractive index becomes spatially inhomogeneous so that in the middle of the beam the light proceeds slower than in the external zone. The result is that the medium behaves like a converging lens. Nonlinear media with these characteristics are nematic liquid crystals (NLC) that have been largely investigated especially using visible laser light [3,4]. They exhibit an extraordinarily high optical nonlinearity, showing self–action phenomena like self–focusing and self–phase modulation [5,6]. However we know much less of these nonlinear materials in the middle infrared spectral region where, nevertheless, the behaviour of NLCs is important in order to develop electro–optical devices, like shutters, polarizers etc, working at these wavelengths. Among all the characteristic parameters of the NLC, the three elastic constants play a prominent role because they characterize all processes of molecular reorientation. In this paper we investigate the self–focusing of a CO_2 laser beam ($\lambda = 10.6$ μm), induced by a film of the NLC E7. By means of an infrared telecamera we observe the self–focusing effect measuring the diameter of the laser beam after it has passed through the NLC film. Both the spatial distribution of the refractive index of the NLC film induced by the incident laser beam and the intensity profile of the laser light on the observation plane are calculated. The theoretical value of the beam diameter is compared with the experimental one. The average value of the elastic constants of the NLC is retrieved as a best fit of the experimental data.

2 Description of the experiment

The experimental setup is shown in Figure 1. Our light source is a c.w. CO_2 laser that operates on the fundamental Gaussian mode at a wavelength of 10.6 μm (line P20). The laser light is polarized along the incident plane (p-polarization) by means of a Brewster window placed inside the laser cavity. A beam attenuator allows us to vary continuously the power of the laser beam. A ZnSe lens of focal length 150 mm focuses the laser light onto a NLC sample. Finally the laser light reaches an infrared pyroelectric telecamera (Spiricon Pyrocam III, model PY–III–C–A) interfaced with a personal computer, which, by means of a software, allows us to perform a detailed analysis of the captured images. The NLC cell consists of two KBr windows separated by a Teflon spacer (150 μm thick) and is filled with a film of the NLC E7. In order to achieve the

[a] e-mail: stefanob@ino.it

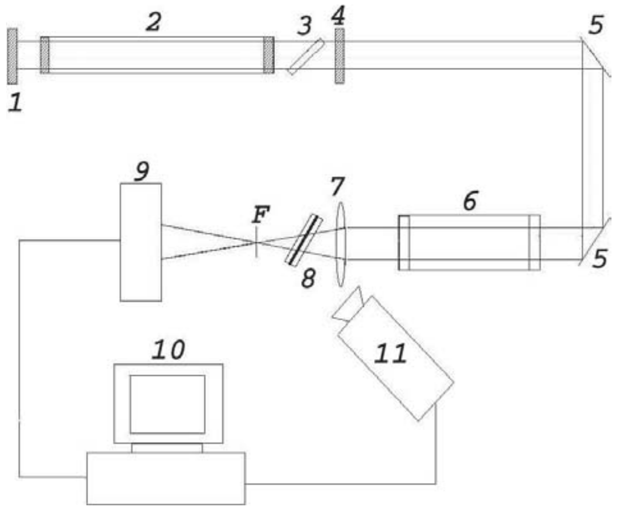

Fig. 1. Experimental apparatus: (1) total reflecting mirror, (2) gas tube, (3) Brewster window, (4) out–coupling mirror, (5) mirrors, (6) beam attenuator, (7) ZnSe lens, (8) liquid crystal cell, (9) spiricon infrared telecamera, (10) personal computer, (11) infrared radiometer.

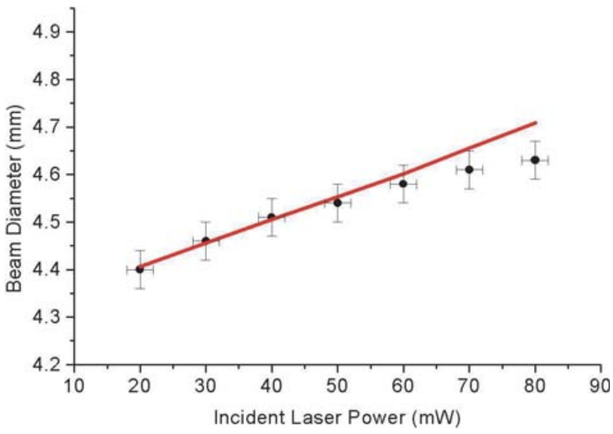

Fig. 2. Comparison between the calculated beam diameter on the observation plane (continuous line) and the experimental one (dots) versus the incident laser power. As the laser power increases the beam diameter grows less rapidly with respect to the theoretical prevision. This is probably due to the local heating of the NLC sample.

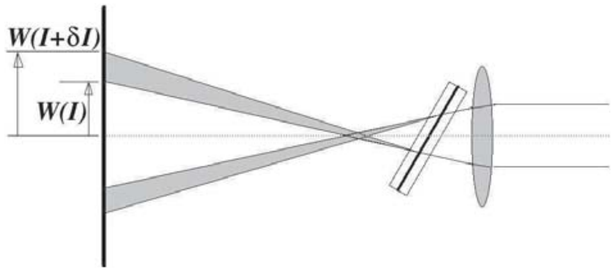

Fig. 3. Scheme of the self–focusing effect in the geometry of our experiment. Because the sample is placed between the lens and its focus the effect give rise to an increasing of the laser beam diameter with the incident laser power.

homeotropic alignment of the NLC molecules the internal surfaces of the windows have been treated with DMOAP (dimethyloctadecyl-3-(trimethoxysilyl) propylammonium chloride) by means of a spin coating technique. The good quality of the alignment has been checked by means of a polarizing microscope. The temperature of the whole NLC cell is maintained at $T = 22 \pm 0.1$ °C, by means of a thermostat and has been measured by means of a thermometer with an accuracy of 0.1 °C. A laser beam that traverses a LC sample gives always rise to a certain local increase of the temperature of the sample, because of the partial absorption of the laser light. To have information about the local temperature of the sample in the zone heated by the laser spot we used an infrared radiometer, which gives us a spatial map of the temperature of the NLC sample. The NLC cell is tilted of $\beta_0 = 30°$ with respect to the light propagation direction $\vec{k}_0$ around an axis perpendicular to the plane of incidence, in order to avoid a threshold effect in the molecular reorientation. The diameter of the beam transmitted by the sample is measured on the detection plane. In Figure 2 we plot the transmitted beam diameter (dotted curve) as a function of the incident laser power. A linear behaviour followed by a saturation is observed as the laser power increases. Because the sample is placed between the lens and its focus this behaviour can be interpreted as a self–focusing phenomenon (see Fig. 3). Each value of the beam diameter has been obtained averaging over 25 different captured frames in order to reduce the error. By means of paraxial ray analysis the diameter and the curvature radius of the laser beam at the entrance plane of the NLC have been estimated to be 1400 μm and -4 cm respectively. It is important to notice that for the laser intensity values used in this experiment we are well below the intensity threshold that leads to self–phase modulation phenomena. Such a threshold has been measured to be $I_{Th} = 36$ W/cm^2. In our experiment the laser maximum intensity reached is of only 5.2 W/cm^2. Some considerations about the response time of the cell are now needed. The measurement of the response time T of the cell has been carried out by the aid of a chopper in order to provide a periodic interruption of the laser beam incident on the NLC. A HgCdTe fast detector, placed in a given point of the observation plane, has been used to record the laser transmitted intensity. The analysis has revealed a transient regime of the order of 200 ms after which the intensity reaches its stationary value.

3 Theoretical interpretation

Liquid crystals are compounds which molecules have an elongated shape. In the nematic phase the molecules tend to align themselves in the same direction giving rise to a strong anisotropic optical medium. It is usual to define a vectorial quantity called the director $\hat{n}(\vec{r})$ which gives the alignment direction of the molecules [7]. The director is

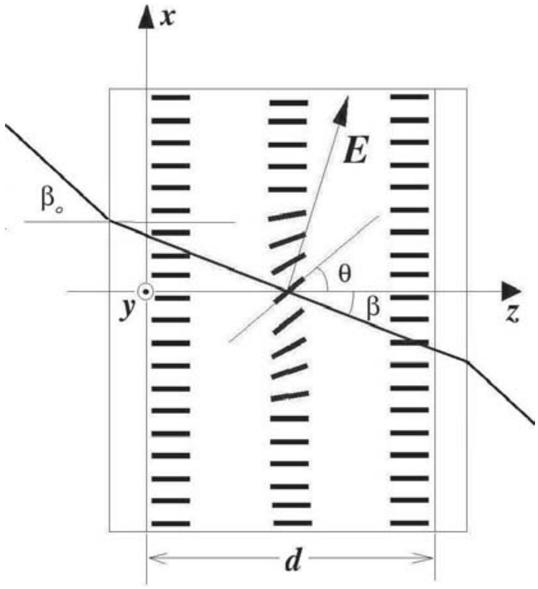

Fig. 4. Scheme showing the geometry of the interaction between the electric field of the electromagnetic wave and the molecules of the NLC. The angle β is the angle formed by the light propagation direction with the normal to the windows, and $\theta(r)$ is the angle formed by the reoriented molecules.

defined as an average over a fluid volume that contains a number of molecules large enough to make meaningful the average process and small enough to make the director vector a function of the position. Let us consider now a linearly polarized electromagnetic wave impinging on the NLC film in the geometry shown in Figure 4. Because of the positive dielectric anisotropy of the medium, the electric field of the wave tends to align the molecules in a direction parallel to the electric field itself, while the elastic torque tends to restore the molecules into the unperturbed condition. In the stationary regime, this leads to an equilibrium condition where the molecules form an angle θ with their unperturbed direction. The incident laser beam has a Gaussian intensity profile described by the expression:

$$I(r) = I_0 \exp\left(-\frac{2r^2}{w_0^2}\right) \tag{1}$$

where w_0 is the beam waist and where the maximum intensity I_0 is connected to the laser power P by the relation $I_o = 2P/(\pi w_0^2)$. In our experimental conditions the light propagates into the NLC and experiences a refractive index provided by the expression (see Ref. [8])

$$n_{eff}(r) = \frac{n_\perp n_\parallel}{\sqrt{n_\parallel^2 \cos^2(\beta+\theta(r)) + n_\perp^2 \sin^2(\beta+\theta(r))}} \tag{2}$$

where β is the angle formed by the wave vector $\vec{k_0}$, inside the liquid crystal, with the window normal and $\theta(r)$ is the reorientation angle due to the influence of the electric field of the electromagnetic wave (see Fig. 4). Here r represents the radial coordinate on the NLC film. In what follows we will use the one–elastic constant approximation, i.e. $K_{11} = K_{22} = K_{33} = K$ [4]. Since the beam diameter and the curvature radius of the laser beam on the NLC film are much greater than the film thickness, the laser wave can be considered as a plane wave and the subsequent theoretical conclusions can be derived (see Ref. [4]). The balance torque equation is given by

$$K\frac{d^2\theta}{dz^2} + \frac{\Delta\epsilon|E_{opt}|^2}{16\pi}\sin 2(\beta+\theta) = 0 \tag{3}$$

where K and $\Delta\epsilon = \epsilon_\parallel - \epsilon_\perp$ are the elastic constant and the dielectric anisotropy of the NLC respectively, $\vec{E}_{opt}$ is the incident electric field and d is the cell thickness. We assume the strong anchoring condition, i.e. the molecules close to the cell windows are strongly anchored to the surface of the substrate and they remains always perpendicular to it so that we can write

$$\theta(z=0) = \theta(z=d) = 0. \tag{4}$$

With these boundary conditions and in the small reorientation angle approximation ($\theta(r) \ll 1$) [4] the solution of equation (3) can be fairly well approximated by the expression [3,4,8]

$$\theta(z,r) = \frac{\Delta\epsilon}{4cK}\frac{2P}{\pi w_0^2}\exp\left(-\frac{2r^2}{w_0^2}\right) \times \frac{\sqrt{n_\parallel^2\cos^2\beta + n_\perp^2\sin^2\beta}}{n_\perp n_\parallel}\sin(2\beta)(dz-z^2). \tag{5}$$

where we taken into account the relationship between the laser intensity I and the optical electric field E_{opt}. Here $\beta+\theta \simeq \beta$ because θ is much smaller than β. The shape of the Gaussian profile of the laser beam has also been considered. Let us introduce the subsequent approximation: we average about z between $z=0$ and $z=d$, [4] replacing the last term in parentheses with $d^2/6$, obtaining

$$\theta(r) = \Gamma\sin(2\beta)P\exp\left(-\frac{2r^2}{w_0^2}\right) \tag{6}$$

where

$$\Gamma = \frac{\Delta\epsilon}{12cK}\frac{d^2}{\pi w_0^2}\frac{\sqrt{n_\parallel^2\cos^2\beta + n_\perp^2\sin^2\beta}}{n_\perp n_\parallel}. \tag{7}$$

Substituting equation (6) into equation (2) we obtain the refractive index seen by the laser wave:

$$n_{eff}(r) = n_\perp n_\parallel\left[n_\parallel^2\cos^2\left(\beta + \Gamma\sin(2\beta)Pe^{-\frac{2r^2}{w_0^2}}\right) + n_\perp^2\sin^2\left(\beta+\Gamma\sin(2\beta)Pe^{-\frac{2r^2}{w_0^2}}\right)\right]^{-1/2}. \tag{8}$$

Here we notice that the refractive index of the NLC depends on the laser power P and on the radial coordinate r. The light impinging on the sample acquires a nonlinear phase shift Φ_{NL} inside the NLC film and then propagates in free space until it reaches the detector plane of the telecamera. The laser beam profile on the observation plane can be described by the Kirchhoff's diffraction integral which takes the form (see Refs. [9–11])

$$I(r_1, Z) = \left(\frac{2\pi}{\lambda Z}\right)^2 I_0 \left| \int_0^\infty r dr J_0(2\pi r r_1/\lambda Z) \times \exp\left(-\frac{2r^2}{w^2}\right) \exp[-i(\phi_D + \phi_{NL})] \right|^2 \quad (9)$$

where

$$\phi_D(r) = k_0 \left(\frac{r^2}{2Z} + \frac{r^2}{2R}\right) \quad (10)$$

$$\phi_{NL}(r) = k_0 n_{eff}(r) d. \quad (11)$$

$k_0 = 2\pi/\lambda$ is the modulus of the wave vector of the incident laser radiation, r is the radial coordinate on the NLC plane, J_0 is the zeroth–order Bessel function, r_1 is the radial coordinate on the observation plane and Z is the distance between the observation plane and the NLC film. R and $2w$ are the curvature radius and the diameter of the laser beam at the entrance plane of the NLC film respectively. ϕ_{NL} is the phase shift due to the nonuniform refractive index profile, caused by the reorientation angle distribution of the NLC molecules. The Kirchhoff's diffraction integral (Eq. (9)) has been numerically solved. We calculated the transmitted radial beam profile on the detector plane, extracting from it the value of the beam diameter. The quantity Γ has been changed in an iterative way in order to obtain a best fit of the experimental data. In Figure 2 we show the comparison between the calculated (continuous line) and the measured beam diameter (dotted curve) versus the laser power for $\Gamma = 0.347$ W^{-1}, while, in Figure 5, we report the radial profile of the reorientation angle for the same value of Γ. It is worth noting that for the reorientation angle the condition $\theta \ll 1$ is verified, confirming that the assumption of the small reorientation approximation is justified. From Figure 2 we see that the calculated beam diameter agrees with the experimental one especially for low laser powers while, for higher values, the beam diameter tends to increase less rapidly. This difference in the behaviour is due to thermal effects which tends to defocus the laser radiation because of the induced local thermal expansion of the NLC. Thus the heating of the NLC produces an antagonist effect to the optical reorientation of the molecular director. As a consequence, the beam diameter increases more slowly than is expected by the theoretical model. From measurements performed by means of the infrared radiometer it results that the spatial distribution of the temperature of the sample in the zone heated by the laser spot has a bell shaped radial profile of approximately 1 mm diameter. In Figure 6 we report the temperature profile of the sample as a function of the radial distance from the center of symmetry for a laser power of 66 mW. In the center of the spot the temperature has raised to 42.5 °C. The local average temperature is $\langle T \rangle = 30$ °C where this value has been obtained as an average both over space and over the range of laser power of interest in our experiment. At $T = 30$ °C and at a wavelength of 10.6 μm, according to reference [4], the values of the extraordinary and ordinary refractive indexes are $n_\parallel = 1.69$ and $n_\perp = 1.49$ respectively. Substituting these values into equation (7) we obtain $K = (0.5 \pm 0.08) \times 10^{-6}$ dyne. This value is comparable to that found in previous experiments. In particular, in reference [12], the bend elastic constant has been obtained exploiting the optically induced Freedericsz transition effect in a different experimental geometry and for $T = 25$ °C, obtaining $K_{33} = 0.7 \times 10^{-6} \pm 0.1 \times 10^{-6}$ dyne.

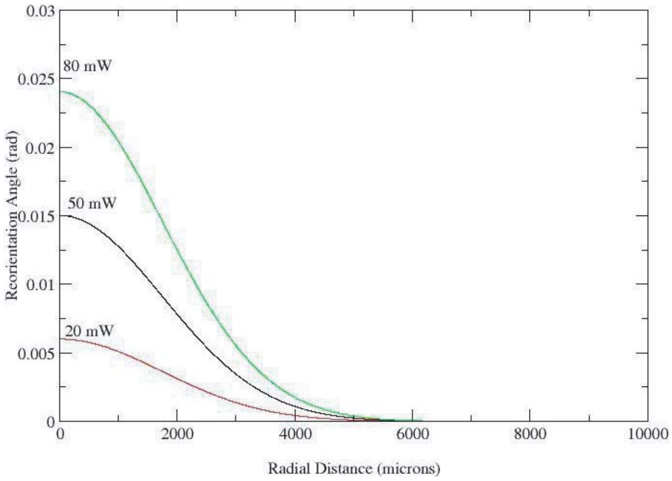

Fig. 5. Plot of the calculated values of the angle $\theta(r)$ formed by the molecular director with respect to the unperturbed molecular axis with respect to the radial coordinate for three different laser powers.

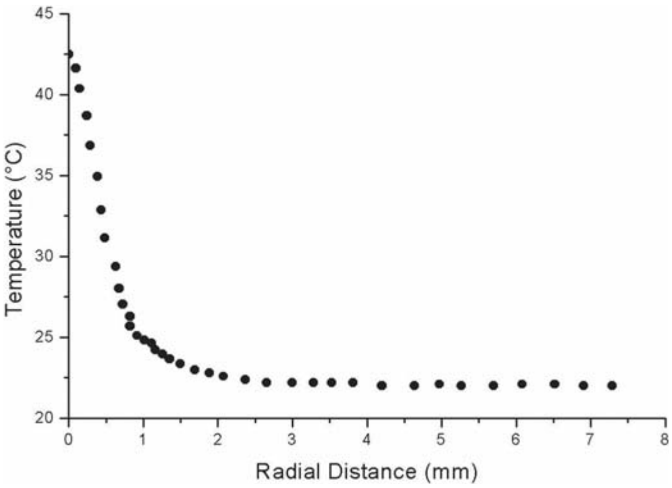

Fig. 6. Temperature profile of the part of the NLC sample heated by the laser spot for a laser power of 66 mW. The temperature profile is bell-shaped and in the central part of the spot the temperature has raised to 42.5 °C.

The discrepancy between this two values can be ascribed to the different temperature at which the two elastic constants have been estimated.

4 Conclusions

The value of the average elastic constant of a particular liquid crystal has been measured by a pure optical method exploiting the self–focusing of a middle infrared laser beam caused by the nonlinearity of the liquid crystal. Self–focusing has been theoretically interpreted as a result of the reorientation of the molecules of the liquid crystal induced by the electric field of the incident wave. The Kirchhoff's diffraction integral has been numerically solved in order to reproduce the intensity profile of the transmitted laser beam on the observation plane. We also considered the heating of the liquid crystal film caused by the partial absorption of the laser light by the liquid crystal, which at this wavelength is not completely negligible. The nonuniform temperature field on the liquid crystal film induced by the laser beam has been measured by a thermographic technique. The application of thermography with this aim is new for this kind of experiments.

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