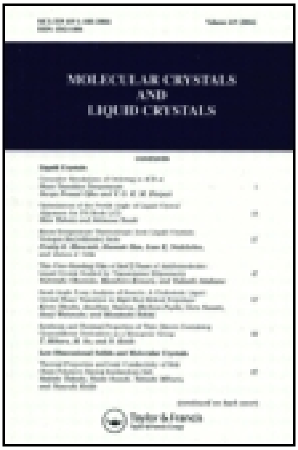

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Characterization of Optical Nonlinearity in Dye-Doped Nematic Liquid Crystal and the Effect of AC Voltage on Its Behavior

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In this paper the nonlinear refractive index, n_2, of an azo dye (Sudan III) doped nematic liquid crystal was measured at low laser power using closed aperture z-scan and moiré deflectometry techniques. The positive sign indicates a self-focusing effect that accrued in the sample. The nonlinear refractive index n_2 is in order of 10^{-6} cm^2/W with z-scan method. This observation is found to be in good agreement with the moiré deflectometry model.

The optical response of the nematic liquid crystals was strongly enhanced by a small amount of dye impurities (Sudan III). Also the effect of external applied AC voltage on the nonlinear refractive is studied.

Keywords: azo-dye; closed-aperature setup; moiré deflectometry; nematic liquid crystal; nonlinear refractive index; z-scan

1. INTRODUCTION

The current interest in materials with nematic liquid-crystalline properties is applied in electro-optical displays, optical storage devices, and nonlinear optics. The massive optical nonlinearity of pure nematic liquid crystals arises from their large refractive index anisotropy coupled with the collective molecular reorientation [1–3]. The optical nonlinearity of nematic liquid crystals can be further enhanced by a small amount of dye impurities [4,5].

The nonlinear optical behavior depends on several factors such as temperature, dye impurities, laser intensity, wavelength, and nematic molecular structure. Therefore, it is interesting to investigate the

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influence of these effective factors on the nonlinear optical properties of the liquid crystals.

In this work, the nonlinear refractive index, n_2, of an azo-dye (Sudan III) doped nematic liquid crystal was measured at low laser power. Two experimental setups are used for this purpose, closed aperture z-scan and moiré deflectometry technique.

In close-aperture test, the sample is simply scanned across the focal point of a 532 nm laser beam past a short-focal-length lens. As the sample passes through the focal point of the beam, the power density changes. Variation in transmitted intensity is related directly to nonlinear optical (NLO) index coefficient [6].

Another method shows when a nonlinear sample is placed in moiré deflectometry setup, the moiré fringe patterns will rotate around the beam center because of self-focusing effects in sample. By measuring this rotation the magnitude and sign of nonlinear refractive index were calculated [7,8].

We found generally good agreement between the values of n_2 determined by z-scan and that from moiré deflectometry. The results indicate that the nonlinear refractive index is positive and in order of 10^{-6} (cm^2/W) at 532 nm by using 10 mW laser beam power where the nonlinear refractive index of pure nematic crystal was in order of 10^{-8} (cm^2/W).

2. EXPERIMENTS

2.1. Materials

In this paper, the nonlinear refractive index of Sudan III doped nematic liquid crystal is measured at 532 nm wavelength. A dichroic diazo dye (Sudan III) was used as the guest material, and 4-trans-4-hexyl cyclohexyl isothiocyanatobenzene (6CHBT) used as the host materials.

The molecular structure of Sudan III and 6CHBT are shown in Figure 1. The dye is dissolved in the nematic liquid crystal with 1% concentration and filled in liquid crystal cells [9].

The homeotropic orientation of the guest and host molecules (perpendicular alignment) was achieved using these specific cells. The thickness of the cell is 12.5 μm whose absorbance in the visible range is negligible small. The temperature of the sample was kept constant at the room temperature.

2.2. Z-Scan Measurement

The z-scan technique was presented in detail in Sheik-bahae *et al*. [6]. This method is based on the investigation of changes in Gaussian

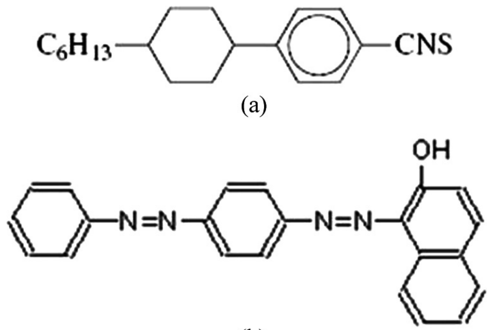

FIGURE 1 Molecular structure of (a) 6CHBT (b) Sudan III.

beam intensity profile in the far field during the moving of the sample through the focal plane. The main advantage of this method is the fact that for the determination of nonlinear optical characteristics. It is only necessary to know transmission data of the investigated sample.

It consists of focusing a beam by a single lens to a narrow waist. The sample was positioned near the beam waist, and the transmittance was measured with a power meter behind an aperture. The sample was moved along the direction of beam propagation, the z-direction, and the transmittance versus sample position data gives information about the nonlinearities. The laser radiation was focused by a 10 cm focal length lens, FL, as shown in Figure 2.

In this section, we present some equations we have used for analysis of nonlinear optical characteristics of colloidal metals. For calculations of nonlinear–optical parameters of the sample, we used equations of the z-scan theory:

$$\Delta T_{p-v} = 0.406\ (1 - s)^{0.25} \Delta\Phi_0 \tag{1}$$

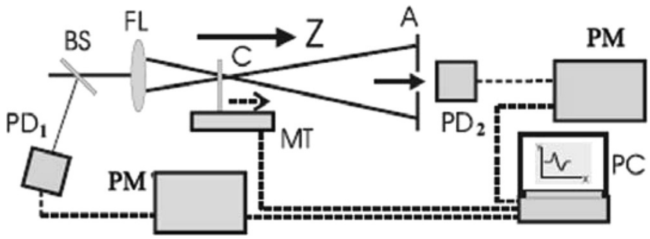

FIGURE 2 Experimental setup of closed-aperture z-scan. FL, focusing lens; A, aperture; PD, power meter; DV, digital voltmeters; BS, beamsplitter; IS, investigated sample; MT, moving table.

and

$$\Delta\Phi_0 = \frac{2\pi L_{eff} n_2 I}{\lambda} \tag{2}$$

where $\Delta T_{p\text{-}v}$ is the normalized difference between peak-to-valley transmission, I is the radiation intensity, s is the transmission of the aperture, and $\Delta\varphi_0$ is the phase distortion of radiation passed through the cell. L_{eff} is the effective length of the sample, and can be determined as follows:

$$L_{eff} = \frac{1 - e^{-\alpha L}}{\alpha} \tag{3}$$

where L is the length of the sample and α is the linear absorption coefficient.

2.3. Moiré Deflectometry Technique

The moiré deflectometry experimental setup is shown in Figure 3. When a sample with nonlinear refractive properties moves between two lenses L_1 and L_2, the location of the focal point will change. Thus the moiré fringes would rotate as their size change. For example a sample with $n_2 > 0$, the self-focusing occurs and the focal point moves far from L_2 so the incident beam on moiré deflectometer converges and the fringes sizes become smaller. Reversely in the case of self-defocusing effect, the focal point moves toward and the incident laser beam diverges, and when the sample is in the focus, the fringes does not rotate. This is the same as the case when the sample is far from the focal point. For far distances from the focus the incident beam irradiance on the sample is very low and rotation is negligible.

The beam deflection angle, which carries information about change in refractive index [10] within the sample, depends on the sample position, so by calculation of ray deflection we can yield to the fringes rotation angle.

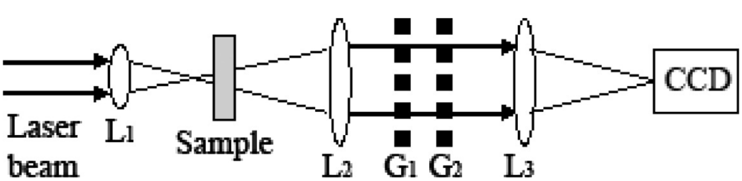

FIGURE 3 Experimental setup of moiré deflectometry. L_1, L_2, L_3 focusing lenses; G_1, G_2 gratings.

For a thin nonlinear medium of thickness L, the parabolic approximation yields a thin spherical lens and its effective focal length is obtained by:

$$f_{eff}(z) = \frac{\pi\omega^4(z)}{4Ln_2P_0} = f_{eff}(0)\left[1 + \frac{z^2}{z_0^2}\right]^2 \tag{4}$$

Using Eq. (4), the nonlinear refractive index relates to moiré fringe rotations [8] and by measuring this rotation n_2 will be obtained.

3. EXPERIMENTAL RESULTS AND DISCUSSION

Z-scan measurements were performed at 532 nm as shown in Figure 4(a), this wavelength is close to the absorption peak of UV-Visible of sample. Linear transmittance for closed-aperture is $s = 0.2$. Photo detector and power meter (Lab-Master, Coherent) placed behind a small aperture records the transmittance intensity. The curve of closed-aperture suggests that the refractive index change is positive, exhibiting a self-focusing effect. In addition, the configuration of this curve shows a symmetric valley-peak, suggesting that the nonlinear absorption is negligible. By using Eqs. (1–3) the nonlinear refractive index will be $8.4 \times 10^{-6}\,\mathrm{cm^2/W}$ in this method.

In the other method, the collimated light in Figure 3 is allowed to fall on the test sample and then propagates through a pair of transmission gratings placed at a distance from each other. Moiré fringe patterns are formed by superimposing one the Talbot images of the first grating on of the second one. The resulting fringe pattern is a map of ray deflections corresponding to the optical properties of the inspected sample.

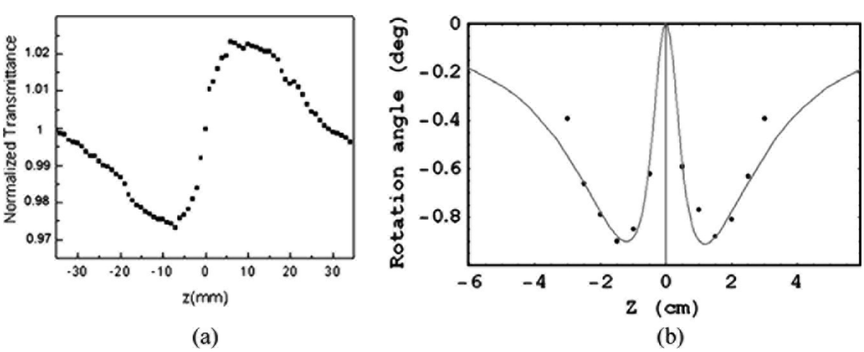

FIGURE 4 (a) Normalized closed-aperture z-scan data. (b) The moiré fringes rotation angle of sample as function of distances from focus.

In our experiment, the best fringes are formed for the forth Talbot [8]. In the moiré deflectometry setup, a 10 mW laser beam is focused by lens L_1 (with 10 cm focal length) and is recollimated by lens L_2 (focal length of 50 cm). The beam intensity in the focus is $1200\,W/cm^2$. Two similar gratings G_1 and G_2 with 80 lines per centimeter are used to construct the moiré fringe pattern. The moiré fringe patterns are projected by lens L_3 placed at the back of the second grating.

By moving the sample from –10 cm to 10 cm and in each step the fringes are recorded. The rotations are measurable in -3 to $+3$ cm and they are very small in the other distances so we couldn't measure the exact rotations in other positions, although the magnitude of n_2 depends on the depth of valleys. Figure 4(b) shows the experimental results and they are fitted to theoretical curve (solid line) from Eq. (4). The best fit yields that the n_2 magnitude is $6.2 \times 10^{-6}\,cm^2/W$.

Although the moiré deflectometry is more complex but the n_2 value is in a good agreement with z-scan method and in both methods, it is in order of $10^{-6}\,cm^2/W$. The experiments for pure nematic crystal indicate that the nonlinear refractive index is in order of $10^{-8}\,cm^2/W$. This fact is obtained by using the z-scan method with the sample that did not dope by dye. So this dye can enhance the nonlinearity. It can be inferred that the n_2 for dye-doped liquid crystal is about two orders of magnitude greater than that for pure liquid crystals.

The experimental measurement of n_2 allows us to determine the approximate value of the third-order nonlinear optical susceptibility $\chi^{(3)}$ as I. C. Khoo showed in [11]:

$$\chi^{(3)}_{esu} = 9.54 n_0^2 n_2 \left(\frac{cm^2}{W}\right) \tag{5}$$

where n_0 is the linear refractive index which is measured by a simple experiment ($n_0 = 1.57$). Using Eq. (5), $\chi^{(3)}_{esu}$ is in order of 10^{-5}. In comparison with the other nonlinear optical materials such as organic metals [12], this material may have a potential application in nonlinear optical devices. The nonlinear refractive index is a very important parameter in designing optical devices. The large nonlinearity in this dye shows that it is a promising candidate for photonic devices and LCD application.

Finally the nonlinear refractive index, measured with an external ac applied voltage. For this purpose, a (0.5 V and 1 kHz) power supplier is used. The change of the dyed liquid crystal nonlinearity, which is the direct impact of the applied voltage on the dye molecules orientation, was not observed for the nonlinear response of the pure liquid crystal. But the nonlinear refractive index of this dye-doped nematic liquid crystal is remarkably reduced. The data reflecting this behavior,

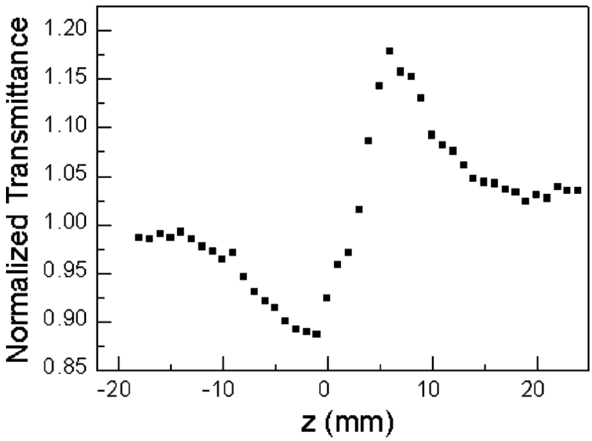

FIGURE 5 Normalized closed-aperture z-scan data with 0.5 V and 1 kHz voltage.

obtained by z-scan technique, is displayed in Figure 5 which yields to $n_2 = 2 \times 10^{-6}$ (cm^2/W).

The orientational distribution and rotational dynamics of the excited dye molecules lead to a nonzero effective torque exerting on the nematic director so in the presence of the applied voltage that is applied in the direction of the liquid crystal director on a homeotropic-aligned cell, the voltage-induced torque can affect the effective torque. Therefore, by increasing the applied voltage the dye molecules can be aligned in the direction of external field and the nonlinearity is decreased.

We choose a low power laser beam to avoid the thermal effects [13] and trans-cis photoisomerization [14].

4. CONCLUSION

In this work, a self-focusing experiment was carried out. The effect of dye on the order of nonlinearity in a nematic hosts was discussed. An enhancement by two orders of magnitude in the nonlinear refractive index of dyed liquid crystals was obtained with respect to pure liquid crystals at low laser intensities (~10 mW). The nonlinear refractive index was calculated by z-scan technique as well as it was studied using moiré fringe patterns too. In both methods the n_2 was in order of 10^{-6} (cm^2/W). Also the nonlinear refractive index was measured with an external ac applied voltage which decreased the optical nonlinearity.

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