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Nonlinear Optical Effects in Nematic Liquid-Crystal Films in the 1.55 μm Spectral Region

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We have studied the optical nonlinearities of nematic liquid crystals in the near IR communication spectral region [1.55 μm]. The origins of the refractive index changes are thermal indexing effect and director axis reorientations. Very large refractive index and phase modulation of several π 's can be generated with mW laser power in micron thick dye-doped nematic liquid crystal films.

Keywords: communication channel wavelength, liquid crystals, near-infrared, refractive index change, laser induced, 1.55 μm .

INTRODUCTION

Refractive index change is the fundamental parameter responsible for the working principles of almost all types of optical devices, including for examples optical logic gate, router, switches, holographic gratings. Nematic liquid crystals (NLC) possess very broadband birefringence ($\Delta n \sim 0.2\text{-}0.5$) and transparency from the 400 nm to 20 microns spectral region. Because of this and other unique physical properties, and acceptable response speeds, NLC's are widely used in various optoelectronics applications [1,2]. Nematic liquid crystals are also highly nonlinear, i.e., they possess very large index changing coefficients n_2 , [defined by the relationship between the index change

Δn and the optical intensity I , $\Delta n = n_2 I$] arising from a variety of field induced effects such as director axis reorientation, thermal and density effects, order parameter modification of the LC's or some dopants [3-9]. In spite of considerable progress achieved so far, most experimental investigations of nonlinearities in doped liquid crystals have been mostly carried out using visible illuminating light. However, as has been shown in some reports, infrared wavelengths are also capable of inducing significant nonlinearities. In particular, functions and effects such as infrared to visible image conversions[10], self-diffraction[11] of CO_2 light or beam amplification by wave mixing[12] and all-optical switching[13] have been reported previously.

The motivation for the work presented here was to explore further the nonlinear capabilities of dye doped liquid crystals at $1.55 \mu\text{m}$, i.e. in the important regime of optical telecommunications wavelengths and consider their applications in photonic devices. In particular, we investigated self-phase modulation in dye doped liquid crystals induced by $1.55 \mu\text{m}$ diode.

LIQUID CRYSTAL INFRARED NONLINEAR OPTICS

In the cw - microsecond temporal regime, the dominant nonlinear optical mechanisms are thermal effect and molecular reorientation. Consider the thermal effect. In the $8\text{-}12 \mu\text{m}$ regime, the absorption constant α of liquid crystals is typically in the range of $40\text{-}100 \text{ cm}^{-1}$; in the mid-IR regime [$\sim 5 \mu\text{m}$], $\alpha \sim$ of 10 cm^{-1} , in the near IR regime, $\alpha \sim 1 \text{ cm}^{-1}$ and in the visible regime, $\alpha < 1 \text{ cm}^{-1}$. There are large variation in the absorption constants among the thousands of existing liquid crystals [1], of course; for actual application, the absorption constants can also be 'adjusted' by using suitable mixtures of liquid crystals or dopants.

For nematic liquid crystals, a change in the temperature will give a positive change in the ordinary refractive index Δn_o , and a negative change in the extraordinary index Δn_e . The onset times τ_{on} and the decay time τ_{off} for the laser induced refractive index change depend on the geometry of interaction such as sample thickness and laser beam size, as well as the thermal conductivities of the liquid crystal and the enclosing materials [1]. A rule of thumb is that for a characteristics interaction length of $15 \mu\text{m}$, τ 's are on the order of

50-100 μsec .

In terms of the nonlinear coefficient n_2 for the index changes, defined by $\Delta n = n_2 I$ [I is the laser intensity in Watts/cm^2], the magnitude of n_2 is on the order of $10^{-4} \text{ cm}^2/\text{Watt}$ for cw laser. For laser pulse shorter than the characteristic response times, e.g. a microsecond laser pulse, n_2 is on the order of $10^{-7} \text{ cm}^2/\text{Watt}$ [1]. The magnitude of n_2 is dependent on the thermal index gradient dn_o/dT and dn_e/dT , as well as the absorption coefficient, and they could therefore vary by several orders of magnitude depending on the t_c of the liquid crystal relative to the experimental ambient temperature, and the concentration of the absorber or dopant used.

The theory and practice of laser induced molecular reorientation in liquid crystals have been standardized for many years [1]. In the liquid crystalline phase, because of the large birefringence and easy susceptibility of the molecular orientation to external fields, the intensity of the laser needed to create substantial reorientation and refractive index change is rather modest. Typically, the reorientation nonlinearity n_2 is about $10^{-4} \text{ cm}^2/\text{Watt}$ for pure nematic liquid crystal. The onset times τ_{on} is inversely proportional to the laser intensity and can be as short as tens of nanosecond for laser intensity on the order of 10^2 MW/cm^2 . The decay time τ_{off} depends on the square of the sample thickness, or laser beam waist [whichever is smaller], and is typically about 30 milliseconds for a 25 micron thick sample. The decay time can be shortened by using liquid crystals of low viscosity and large elastic constants.

EXPERIMENTS

We employed a simple self-phase modulation technique to measure the intensity dependent refractive index coefficient of various nematic liquid crystalline films. Fig. 1 shows the experimental set-up for self-phase modulation effect. The nonlinear phase shift induced by a 1.55 micron laser is read by a separate beam. The 1.55 μm pump beam originating from a diode laser with the power of approximately 7 mW was focussed via a 5 cm focal length lens to a spot diameter of about 150 μm on the liquid crystal cell. A He-Ne probe beam was directed via a beam splitter along the path of the infrared beam and its

1 mm diameter beam was focussed by the same lens to probe the pump beam induced phase modulation effect. A liquid crystal cell was mounted on a z-stage and its position adjusted to coincide with the focal plane of the lens

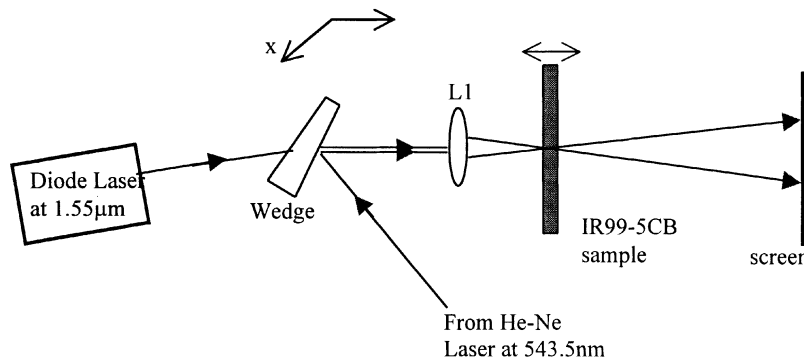


FIGURE 1. Experimental Set-up

Image of the infrared, and the He-Ne beam, emerging from the liquid crystal cell was visualised on a screen placed behind a sample. As the cell's position was scanned through the focal point (along z direction) any changes in the output beams shape were monitored. Initially, both the infrared and the He-Ne laser beams were p-polarised and their incidence normal to the cell's surface. The cell could be rotated versus z -axis. Two positions of the cell—“upright” (director long y axis) and then rotated by 90° (director along x axis) were considered.

For the purpose of our study we prepared several planar samples. We used nematic liquid crystal 5CB or E7 (EM chemicals) doped with different, infrared sensitive dyes. Planar samples were made by putting dye-doped liquid crystal into 20 to 200 μm thick planar-alignment made with plane glass windows, in which the inside surfaces were coated with a thin and rubbed PVA (Polyvinylalcohol, Kodak) layer. Some samples are made with ITO coated galss windows which also exhibit laser heating effect. Table 1 provides the details of all the samples we used and their absorption coefficients at 1.55 μm .

Table 1.

Sample	Thickness [μm]	Dye concentration	Liquid crystal	Absorption coefficient at 1.55 μm [cm^{-1}]	Number of SPM rings observed
1	25	18% azobenzene (ALC)	E7	9.5	4
2a**	20	20% IR99 dye	5CB	4	11
2b**	20	pure	5CB	4	11
3	50	0.5% A118 dye	5CB	14.3	6
4	50	0.5% A156 dye	5CB	4.7	6
5	60	0.4% Epolite 125 dye	5CB	11	7
6	200	no dye (pure)	5CB	1	3

**** Sample made with ITO coated windows.**

Phase modulation effect on a beam profile originating from a thermal effect

Strong self-induced modulation effect at 1.55 μm could be easily observed in all the samples. It manifested itself, as expected, as a modification of the probe beam profile, namely a pattern of rings with bright and dark circles. The ring pattern changed depending on the z-position of the liquid crystal cell, namely its position versus the focal point. For example, the pattern could have had a large dark circle in the middle surrounded by several bright and dark rings or a bright centre surrounded by dark and bright rings [1,14].

Self-induced modulation effect was observed in all the samples [except the pure 5CB sample] at normal incidence of both beams. In the case of the IR99 doped sample, we observed that similar phase modulation effect can be induced if the cell is made with ITO coated

windows. We attribute the effect to laser heating of the ITO coating, as similar effects can be observed in samples made with pure 5CB [i.e. without any dye doping] and ITO coated windows.

Because 5CB absorbs very little in the $1.55\text{ }\mu\text{m}$ area, in general there is no observable thermal effect for samples prepared with plane glass windows. One means of creating a refractive index change in these non-absorbing samples is orientational effect. To illustrate this effect, which is now very well standardized [1], we employed a $200\text{ }\mu\text{m}$ thick sample of pure 5CB. The sample has to be tilted (inclined) at 50° versus incident [extraordinary-polarized] beam. In general, we observed that the magnitude of phase modulation is smaller than the thermal effects obtained in the dye-doped samples or the sample made with ITO-coated windows.

Figure 2 show a He-Ne beam profile in when the infrared beam is on (figure 2a) and then infrared beam switched off (figure 2b). Typical dimensions of a the outer diameter of a ring pattern was approximately 12 cm with the infrared beam on and 1 cm in diameter when the infrared beam is switched off, i.e. the divergence of the probe beam was increased by 12 fold. These measurements were taken with screen to sample separation of 11 cm and in sample doped with A118 dye.

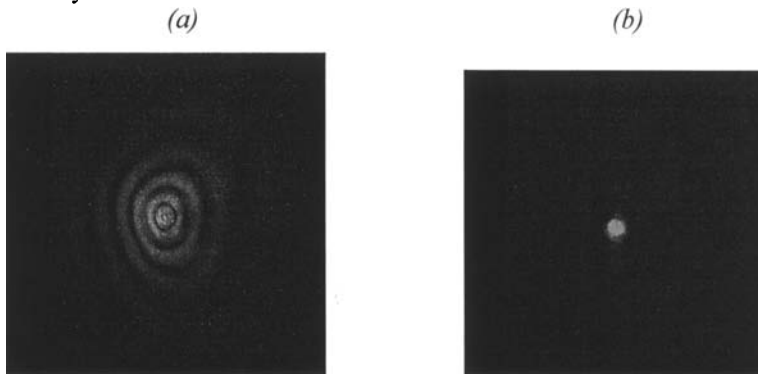


FIGURE 2. He-Ne probe beam profile (a) when $1.55\text{ }\mu\text{m}$ pump beam is on (b) pump beam off.

In all the samples, a typical pattern of modulated green He-Ne probe included several rings, namely from 3 to 11 rings depending on the sample (Table 1). The strength of refractive index change can, in

fact, be deduced from such a ring structure, according to the equation:

$$\phi = 2\pi/\lambda * \Delta n * L$$

where L is the thickness of the film and the phase change ϕ can be estimated as $\phi = N$ (the number of fringes) $* \pi$.

Consider the sample made of 5CB doped with 0.4% [by weight] of Epolite 125 dye. We observed 7 rings in a 60 micron thick cell. This gives $\Delta n \sim 0.032$. Since the intensity used is about 30 W/cm^2 , this gives an index coefficient of 1.04×10^{-3} . The typical order of magnitude of thermal index coefficient of nematic liquid crystal is 10×10^{-4} ; the larger value obtained here for 5CB is simply due to the fact that the measurement was conducted at room temperature [$\sim 24^\circ \text{C}$]. The laser heating raise the temperature of the cell to close to the T_c of 5CB [$\sim 35^\circ \text{C}$] when the thermal index coefficient increases by more than an order of magnitude [1].

We have investigated the speed of modulating the probe beam profile when the infrared beam was switched on and off. A pinhole was set up with a size equal to the size of a probe beam spot in the absence of the infrared light, with a detector was placed behind it. The amount of light passing through the pinhole decreased as the infrared light made the single intense probe beam spot to expand into a ring pattern. Although being limited by the response of the mechanical shutter that was used to block the $1.55 \mu\text{m}$ beam and by the response of the power meter, we were able to establish the upper limit of response times. Fig. 3 shows a typical oscilloscope trace of the on-axis probe transmission as the pump beam is turned on, showing the dynamics of the defocusing effect.

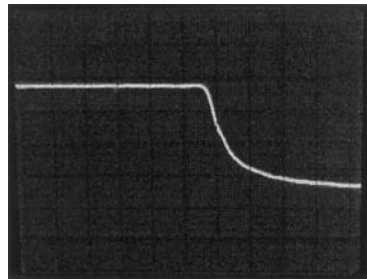


FIGURE 3. Oscilloscope trace of the transmitted probe beam on-axis power.

We determined that the formation of the ring pattern typically took 10-15 ms to build-up in all dye-doped samples we studied. In most of the samples, except the one doped with IR99, the decay of the ring pattern when the infrared beam was blocked, again took up to 15 ms. In case of the IR99 doped sample, this decay time was much longer, up to 100 ms. The difference is most likely due to the different thermal conductivity and diffusion in the liquid crystals and ITO layers. As expected, the orientationally induced phase modulation observed in pure, undoped 5CB sample occurred on much longer scale, namely both the build-up and decay being in the range of 1 second. Such timescale is typical for an effect originating from an orientational nonlinearity.

CONCLUSION

In conclusion, we have investigated the near IR nonlinear optical properties of nematic liquid crystal films. Typical nonlinear index coefficient obtained in these materials are in the range of 10^{-4} to 10^{-3} . A phase shift of several π 's can be induced in microns thick sample with mW laser power. Typical response speed is in the few milliseconds range, and can be as fast as tens of microseconds for shorter (smaller) thermal diffusion length [1]. Clearly, these near IR nonlinear NLC films can be employed for switching, image processing and other nonlinear optical applications that have been demonstrated in the visible regime [1-10, 15,16]. Because of the fluid nature of liquid crystals, they can also be made in planar waveguide[17]or fiber structures for a variety of applications in the 1.55 micron area.

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