Nonlinear absorption and refraction of linearly polarized nanosecond laser radiation by liquid crystals in the transient regime

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ABSTRACT

This work aims at understanding how the liquid crystal device geometry affects its transient nonlinear response to 6-9 ns laser radiation, 10-Hz repetition rate. Results reported here are from Z-scan measurements at 0.532-µm laser wavelength (beam diameters at focus were ~35 - 50-µm) of two pure liquid crystals with “rod-like” molecules of similar chemical composition (C₉H₁₁ alkyl chain, cyano-biphenyl), but distinguished by conformal differences, i.e., 5CB and chiral CB15.

Two types of liquid crystal layers were selected: (1) isotropic liquid (“molecular rods” are chaotically oriented), and (2) planar nematic with “molecular rod” orientation parallel to the cell surface.

(1) For isotropic 5CB and CB15, the transient value of n₂ for ~35 - 50-µm beam diameter at 0.532-µm is found to be several times larger than that of CS₂, with the same positive sign. The transient value of nonlinearity for the fixed laser pulse duration may be controlled by proper irradiation geometry (beam waist radius) triggering different mechanisms of nonlinearity (orientational and thermal-densities) with opposite sign and different build-up times. The nonlinear absorption coefficient (cm/GW) at 0.532-µm of both 5CB and CB15 in the isotropic state was found to depend on the incident intensity and cell thickness. Possible mechanisms of nonlinear absorption and energy dissipation are discussed.

(2) For planar nematic 5CB cumulative negative nonlinearity was observed at 10-Hz repetition rate operation (0.532-µm) for the linear polarization parallel to the orientation of liquid crystal molecules (for the case when no reorientation of the liquid crystal molecules is expected to take place). Values for n₂ more than an order of magnitude larger than for CS₂ were obtained, depending on the incident intensity. At the highest intensity, this nonlinearity manifests itself in form of concentric elliptical diffraction rings with major axis perpendicular to the incident polarization.

Keywords: Z-scan, nanosecond laser, 5CB, CB15, nematic and isotropic liquid crystals

1. INTRODUCTION

Studies of Kerr-like optical nonlinearities and nonlinear transmission changes in liquid crystals (LCs), both in the aligned (oriented) nematic (see, e.g., Figure 1.a for the case of planar alignment), and isotropic state (Figure 1.b) had shown that LCs can be promising materials for optical switching and optical power limiting applications, see, e.g. reviews 1-4. Importance of deeper understanding of nonlinear optical response of LC materials was caused not only by their strong nonlinearities but also by the possibility to use them as a solvent for different chromophores, increasing the nonlinearities of the combined systems, for LCs see, e.g. References 5-8.

As to the ordered nematic state, giant refractive optical nonlinearity of nematic LCs with “rod-like” molecules was discovered. For CW-laser operation this orientational optical nonlinearity was found to be nine orders of magnitude higher than the nonlinearity of such a well-known self-focusing medium as CS₂. For practical applications, this nonlinearity suffers from a slow response time (from submilliseconds up to seconds). Using high-intensity, short-pulse laser radiation may speed up orientational nonlinearity even of oriented nematics dramatically, although at the expense of its magnitude. Even for nanosecond pulse duration diffraction rings of the orientational self-focusing in Gaussian beams were observed.

Isotropic LCs are very attractive because of their excellent transparency even for large beam paths (up to tens of cm, opening an opportunity for use as nonlinear materials in fibers) in difference to nematics which scatter light in all unoriented layers (orientation is difficult to maintain for layer thickness above ~200-µm). For the isotropic LCs, the value of n₂ was measured to be more than ~100 times in excess of CS₂ with a dependence on the temperature build-up time t_w of the order of several tens to hundreds of nanoseconds. If the input pulse is short (pulse duration t_p << t_w), then the optical
field-induced refractive index changes in first approximation\(^{19}\) \(\Delta n(t) \equiv 1/2 \int_{-\infty}^{\infty} n_2 |E(t')|^2 dt'/T_n\). For 6-9-ns pulse duration, the orientational nonlinear refraction is transient and \((n_2)_{\text{transient}}\) should have a smaller value than steady-state \(n_2\) of isotropic LCs.\(^{12-18}\) Both \(n_2\) and \(T_n\) due to molecular reorientation, are proportional to \((T-T_c)^{-1}\), where \(T_c\) is the nematic/isotropic phase transition temperature. As a result, \(\Delta n(t)\) and \((n_2)_{\text{transient}}\) are nearly independent of the temperature.\(^{19}\)

A large number of experiments have been reported since the 1970-80s on the nonlinear optical response of LCs to short-pulse irradiation, both in isotropic-state and ordered nematics.\(^{14,7,8,10-19,21-36}\) It should be noted that electronic nonlinear susceptibility \(\chi^{(2)}\) of LCs is only \(-10^{-14}\) esu and its influence is negligible. With regard to nonlinear absorption, it is by now a widely held notion that heating of pure LC materials, and various thermal effects connected with it, are driven at 0.532-\(\mu\)m by two-photon absorption and concurrent or subsequent excited-state absorption.\(^{26-39}\) The conversion to thermal energy is explained by the efficient decay of excited states through radiationless-recombination channels. For nematic LCs, the two-photon absorption coefficient depends on the incident polarization.\(^{21,26}\)

As to thermal nonlinear refraction, several nanosecond mode, with beam-waist diameter of some tens of micrometers (which is the case of optical power limiting devices and Z-scan measurements) transient thermal-density nonlinearity with negative sign, for small beam radius, can compensate the effects of positive-sign transient orientational nonlinearity, or even dominate. The build-up time of thermal-density nonlinearity \((\partial n/\partial \rho)_{T}\Delta n\) \(\approx r/V_s\) (where \(\rho\) is the density, \(r_0\) is the beam radius and \(V_s\) is the velocity of sound) can be close to the laser pulse duration. For \(t_n \gg t_0\) the thermal nonlinearity will not develop for the pulse-duration time. In Reference 40, the value of transient thermal nonlinear refraction of toluene with dye dissolved in it, for \(t_0 = 5\) ns, was found experimentally to diminish with increasing \(r_0\) from 9 to 32-\(\mu\)m. It was also shown there, that thermo-optical effects \(\Delta n = (\partial n/\partial T)_{\rho}\Delta T\) for \(r_0 = 32-\mu\)m are more significant for pulses longer than \(\approx 100\) ns. As to LCs with strong positive-sign, orientational nonlinearity, one can control the refractive nonlinear response of LCs and its sign by the proper choice of beam-waist size.

The aims of our investigation are as follows:

- to understand how LC device geometry (isotropic or oriented planar nematic at various layer thicknesses) affects its transient nonlinear optical response to nanosecond laser radiation of standard commercial lasers and standard optical power-limiting spot sizes (35 - 50-\(\mu\)m beam diameter).

1. For isotropic LCs, the transient nonlinear refractive index \(n_2\) in several-nanosecond time span (for the time interval much smaller than orientational relaxation time) was measured using Z-scan method for beam diameters \(35 - 50\mu\)m.
2. For oriented planar nematic layers, the data of \(n_2\) measurements for polarization of laser light parallel to the orientation of LC molecular "rods" were compared with the isotropic case and with results of References 30-35.

- to understand the reason for the difference of more than two orders of magnitude of the nonlinear absorption coefficients for picosecond and nanosecond irradiation of LCs (both isotropic and planar nematics) observed in References 30,31.

For this purpose nonlinear absorption coefficient \(\beta\) measurements for LC layers with different thickness (both isotropic and nematics) were carried out.

- to understand existing discrepancies between literature values for both the magnitude and the sign of the 0.532-\(\mu\)m, transient, nonlinear refraction of isotropic-state 5CB and chiral CB15 (same chemical composition) for nanosecond excitation.

**2. LIQUID CRYSTAL MATERIAL AND CELL CHARACTERIZATION**

Nematic LCs consist of rod-like molecules with their long, molecular axis oriented predominantly in one direction (director). Above the nematic/isotropic liquid phase transition molecular orientation is chaotic and the LC behaves optically as an isotropic liquid (Figure 1). We used the well known LCs 5CB (K15) 4-cyano-4'-n-pentylbiphenyl, and CB15 (4'-(2-methylbutyl)-4-biphenylcarbonitrile which in difference to 5CB in its pentyl chain contains a chiral carbon (Figure 2). LCs were supplied by BDH. Degassed 5CB served as sample material having been filtered through a 5-\(\mu\)m filter in a clean room, prior to filling of the cell under regular laboratory conditions. Samples of 5CB were kept at elevated temperatures above nematic/isotropic phase transition \((T_c = 35.3^\circ\)C) under control of an Instec heater control unit. CB15 is isotropic at room
temperature. Spectroscopic quartz cells of 0.5, 1 and 2 mm beam pathlength (NSG Precision Cells, Inc.) were used in the experiments with LCs in the isotropic state and with CS\textsubscript{2} for calibration (99.9\% HPLC grade, Sigma-Aldrich).

Figure 1. a) Planar oriented nematic LC layer ; b) Isotropic LC.

Figure 2. Space-filling representation of 5CB (left) and CB15 (right) molecules.

"Thin" cells were made from glass plates with size ~2.5 cm x 2.5 cm. Mylar spacers of different thickness (25, 50, 105 and 125-\mu m) were used between two glass plates. For LCs in the isotropic state we used Fisher Finest Premium microscopic slides of 1-mm thickness.

For oriented nematic layers of 5CB a planar alignment (Figure 1,a) was used at room temperature. To align LC molecules in layer thickness up to 125-\mu m uniformly in one direction, we used spin-coated, rubbed polyimide glass plates (sodolime or borosilicate with the same dimensions as Fisher slides). Coating and rubbing were carried out in a clean room, but making the cells and filling them with the LCs – under ordinary laboratory conditions. Devcon 5-Minute epoxy was used for sealing the thin cells.

3. EXPERIMENTAL

3.1. Experimental set up

In the Z-scan method\textsuperscript{41-45} for measuring nonlinear absorption and refraction, a sample is scanned along the optical, Z-axis through the focus of a lens, while the far-field transmission is recorded as a function of sample position. The nonlinear absorption coefficient is derived by collecting light from the whole beam (open aperture Z-scan). For determination of the nonlinear refraction, a small aperture is used in front of the detector, admitting only rays close to the Z-axis (closed aperture Z-scan). The ratio of closed-aperture measurements divided by open-aperture measurements yields the "Z-scan curve" for determination of nonlinear refraction.

It should be noted that for pulsed, solid-state lasers with beam quality and divergence far from the diffraction limit, the Z-scan method, simple in principle, requires great care in ensuring reliable results\textsuperscript{41}, foremost in assuring verifiable beam quality and precise measurement of the beam caustic near focus\textsuperscript{64-47}. Pulse-to-pulse fluctuations in laser radiation (both in energy, pulse duration and/or in beam diameter) tend to make the Z-scan method, straightforward as it is for CW, power-stabilized laser sources, a nontrivial task for pulsed lasers. As the accuracy of the Z-scan method critically depends on precise knowledge of the waist size, and precise calculation method should include the beam profile, we carried out before and after each experiment, under identical experimental conditions, also Z-scan measurements for CS\textsubscript{2} whose well-known nonlinear refractive index n\textsubscript{2} \approx 1.2 \times 10^{-14} \text{ esu}\textsuperscript{2} permitted to evaluate the beam-spot size in the focus.

The experimental setup used is shown in Figure 3. A frequency-doubled, Q-switched (6 – 9 ns pulse duration at Gaussian FWHM) Nd:YAG laser (Quantel International (Continuum) model YG-682S-10), using a feedback-stabilized, injection-seeded, single-longitudinal-mode, unstable resonator with Gaussian cavity end mirror\textsuperscript{48} and two, single-pass amplifiers, provided, at 10-Hz repetition rate, an output beam-spot diameter of \sim 8 mm at 0.532 \mu m.
To optimize beam quality, a 1.5-mm-diameter Teflon aperture $T_A$ was placed in the central part of the beam\(^4\), such that an Airy spot at a reasonable distance from the Teflon aperture (using mirrors $M$) was formed. A converging, 9.5 or 10.5-cm focal-length lens $L$ was placed at 4 m from the Teflon aperture. To remove far-field side lobes from the Teflon aperture, a 2.75-mm-diameter, metal aperture $A$ was placed in front of the lens. The same aperture $A$ was placed at the reference channel. For maintaining a linear incident polarization and changing the input-pulse energy to the sample, a Glan-prism-polarizer and a quartz $\lambda/2$ waveplate combination was placed in the beam.

![Experimental setup diagram](image)

Figure 3. Experimental set up.

Signal $D_1$ and reference $D_2$ pyroelectric joulemeters (Gentec ED-100A with amplifier EDX-1, vendor recalibrated) or fast photodetectors (Hamamatsu S1722-01 and Motorola MRD510, respectively) were connected to a digitizing oscilloscope (Tektronix TDS 640). An AT-GPIB/TNT board from National Instruments was used for data acquisition and processing. Laser pulse shape was monitored by a sub-ns-risetime Motorola MRD510 photodiode $D$, connected to a Hewlett Packard HP 54111D digitizing oscilloscope.

For closed-aperture Z-scan measurements, an aperture $A'$ was placed in front of $D_1$. Sometimes the entire photodetector was used as the aperture. For open-aperture measurements, we used an additional lens in front of $D_1$ to collect all laser radiation leaving the sample. The samples were placed on an Aerotech mechanical translation stage controlled by an Unidex 1 motion controller.

To measure the average beam waist\(^4\) produced by the focusing lens $L$ and traveled along by the sample during the experiment, an 8-bit, WinCam CCD-beam analyzer by Merchantek (pixel size 8.3 $\mu$m x 8.6 $\mu$m) was used in a separate, low-intensity, equivalent channel. Our measurements\(^7\) show Gaussian spatial distribution of the laser-beam intensity at the lens and Gaussian temporal pulse shape.

The same CCD-camera was used also for acquisition and analysis of far-field patterns during the Z-scan. For adjusting the size of the pattern to the size of the sensor, an additional lens was used between the sample and the camera. Some far-field patterns were recorded from a screen by a standard video camera/digitizer combination, permitting the recording of each pulse.

The accuracy of measurements of $n_2$ and $\beta$ is estimated to be $\pm 20\%$ dominated by the uncertainty of the irradiance.

### 3.2. Evaluation of $n_2$ and $\beta$ from the Z-scan data

The nonlinear refractive index $n_2$ and the nonlinear refractive coefficient $\gamma$\(^9\) are defined by $n = n_0 + 1/2n_2 |E|^2$ and $n = n_0 + \gamma l$. Here $n$ is the total refractive index of the material, and $n_0$ is the linear refractive index, $E$ is the (complex) optical electric field amplitude inside the material, $l$ is the optical intensity. Conversion between $n_2$ and $\gamma$ is made via $n_2[\text{esu}] = (cn_o/40\pi)\gamma[m^2/W]$, where $c$ is the light speed (m/s) in vacuum.
The values of $\gamma$ and $n_2$ can be calculated from the Z-scan curves using the following formulas\textsuperscript{41-42}:

$$\Delta T = 0.406l \Delta \Phi (1 - S^{0.25}),$$  \hspace{1cm} (1)

where $\Delta T$ is the difference between the maximum value of the normalized transmittance $T$ and its minimum value, $\Delta \Phi$ is the nonlinear phase shift on axis during $Z$-scan, $S$ is the linear transmittance of the aperture for a Gaussian input beam ($S = 1 - \exp(-2r_2^2/R_0^2)$), where $r_0$ is the aperture radius, $R_0$ is the beam radius at the place of aperture in the linear regime.

Nonlinear phase shift $\Delta \Phi = 2\pi \lambda^{-1} \gamma I L$, where $I$ is the peak axial intensity at the waist, assuming a temporally and spatially Gaussian-shaped pulse, $L$ is the sample length.

We define intensity $I(r,t) = 2E(\pi \chi^2 r_0^2)^{1/2}\exp(-2r^2/r_0^2 - t^2)$.

Here $E$ is the total energy, the pulse-length full width at half maximum (FWHM) $t_0 = 2\tau(\ln 2)^{1/2}$, $r_0$ is the beam spot-radius at $1/e^2$ level. Using these pulse and beam parameters, $I_{\text{peak}} \approx 2E/(t_0^2\pi r_0^2)$.

(2')

As to nonlinear absorption, we suppose that two-photon absorption\textsuperscript{51} is the principal loss mechanism. The fundamental equation describing this intensity loss with depth $z$ is

$$dI/dz = -\beta I^2.$$  \hspace{1cm} (3)

The solution to (3) is

$$I(z) = I(0)/(1 + \beta I(0)z), \text{ or } z = L, \text{ at } T(L) = 1 + \beta I(0)L = 1 + Q.$$  \hspace{1cm} (4)

Here $T$ is the sample transmittance, $T = 1 - T_{\text{abs}}$, where $T_{\text{abs}}$ is obtained from the open-aperture $Z$-scan curve. For a Gaussian profile

$$T(L) = 2Q^{-1}\pi^{1/2} \int_0^\infty \ln(1 + Qe^{-2x^2})dx.$$  \hspace{1cm} (5)

Expansion of the logarithmic function yields a linear relationship (valid for small $Q$) between $T(L)$ and $I(0)$ where the slope is proportional to the nonlinear coefficient $\beta$:

$$T(L) \approx 1 + 2^{-3/2}\beta I(0)L, \text{ and } T_{\text{abs}} \approx -2^{3/2}\beta I(0)L.$$  \hspace{1cm} (6)

The plots of (5), $T^{-1}(Q)$ and $1 - T(Q)$ are presented at Figure 4, a, b. In our calculations of $\beta$ in this paper we used relation (5) for $T^{-1}(Q)$ in difference to paper\textsuperscript{50} where approximation by formula (6) was used.

![Figure 4. Dependence of $T^{-1}$ (a) and $(1 - T)$ (b) on $Q$.](image)

4. NONLINEAR REFRACTION AND ABSORPTION MEASUREMENTS: RESULTS

4.1. Isotropic liquid crystals

Results from Z-scan measurements of 2-mm 5CB samples in the isotropic state at 45°C for $l = 0.66$ GW/cm$^2$ are presented in Figure 6(a-c). For comparison, Z-scan curves for CS$_2$ with 1 and 2-mm cell thickness are shown also for the same
experimental conditions (Figure 5, d) for $I_1 = 1.4 \text{ GW/cm}^2$ and $I_2 = 1.2 \text{ GW/cm}^2$. From data in Figure 5, c we derive $\gamma(\text{5CB}) = 3.7 \gamma(\text{CS}_2)$ and $n_2(\text{5CB}) = 3.7 n_2(\text{CS}_2)$.

Over the temperature interval 35.6 - 50 °C, the value of $n_2(\text{5CB})$ changed little; it varied from 2.5 to 3.7 $n_2(\text{CS}_2)$.

Typical Z-scan curves for 2-mm, CB15 cells at room temperature are presented in Figure 6 (a-e) for $I = 0.74 \text{ GW/cm}^2$. For comparison, a Z-scan curve for CS$_2$ with 2-mm thickness is presented as well (Figure 6, d) for $I = 0.73 \text{ GW/cm}^2$. The results from these curves yield the following values: $\gamma(\text{CB15}) = 2.6 \gamma(\text{CS}_2)$ and $n_2(\text{CB15}) = 2.5 n_2(\text{CS}_2)$.

In both cases, isotropic 5CB and CB15, we observed self-focusing (converging lens).

Figure 5. Z-scan curves for 5CB: a – open aperture; b – closed aperture; c – closed/open aperture; d – closed aperture for CS$_2$. (S=0.1).

Figure 6. Z-scan curves for CB15: a – open aperture; b – closed aperture; c – closed/open aperture; d – closed aperture for CS$_2$. (S=0.1).
The main results of β-measurements are as follows:

(1) a remarkably large value of β is found for “thin” layers (e.g., for CB15, β ~ 100 - 150 cm/GW for 25-125-μm cells at I ~ 0.7 - 0.8 GW/cm²) in comparison with that from “thick” samples (β ~ 4.5 - 30 cm/GW for 1-2-mm cells of CB15 and 5CB at 45°C at I ~ 0.2 - 0.9 GW/cm²). Typical curves for β versus L for isotropic CB15 are presented in Figure 7.a.

(2) a dependence of β (cm/GW) on intensity (Figure 7.b).

(3) for isotropic 5CB near the nematic/isotropic phase transition, β values fluctuate by up to a factor of ~6. Five degrees above the phase transition up to 50°C, β does not vary beyond experimental error.

An explanation of the dependence of β on L may not rest on the argument of a Rayleigh length shorter than the sample pathlength. Our measurements of the beam caustic near focus47 show the 1/e² beam radius r� to grow to twice its focal-waist value over a length l = 7.5 mm. Both “thin” and “thick” cells used in this experiment were shorter than this Rayleigh length. In addition, for a sample larger than the Rayleigh length, an open aperture Z-scan curve must be flat-bottomed45.

![Graphs](image_url)

**Figure 7.** Nonlinear absorption coefficient measurements: (a) Dependence of β (CB15) on layer thickness at constant incident intensity; (b) Dependence of β of 5CB on the incident intensity (2-mm cell).

### 4.2. Planar nematics

To take into account the well-known nonlinear-absorption anisotropy in the nematic phase, i.e. nonlinear absorption being larger for incident polarization parallel to the nematic director and weaker for perpendicular polarization21,26,30, orientation-dependent measurements were carried out. Figure 8 shows the results from open (a) and closed (b) aperture Z-scans for a 5CB film of 105-μm thickness, irradiated by a parallel-polarization (I = 0.36 GW/cm²) pulse. Results for the ratio of closed-aperture data by open-aperture data are presented in Figure 8, c. For comparison, a closed-aperture Z-scan for CS₂ is shown in Figure 8,d (I = 1.2 GW/cm²). In all cases of Figure 8, S=0.5.

The results of calculations are as follows: γ = -18.5 γ(Cs₂). Evaluation of β yields a value of ~ 126 cm/GW. These large values by themselves are remarkable, but so is the sign of the nonlinear refraction coefficient (negative). These data are in good agreement with those of References30,32-35, where also a negative nonlinear refraction coefficient was observed for an oriented thin layer of 5CB and parallel-polarized laser light.
Figure 8. Z-scan curves for planar nematic (a-c), CS$_2$ (d) and the geometry of irradiation (e).

5. CCD-IMAGING IN THE FAR-FIELD

Z-scan imaging has recently gained broad acceptance, e.g., References $^{52-53}$, because of the added, spatial information offered by this approach.

In the following, far-field CCD-images are presented for the sample-cell positioned each time in the lens focus (for CB15, 5CB, and CS$_2$) and the camera viewing, at constant position, the beam through a neutral-density filter. This filter, together with the window protecting the CCD-matrix, introduces the fringes and other constant patterns in the images below. In Figure 9, far-field beam patterns at the same incident intensity are depicted for: a) no sample in the beam; b) 2-mm cell of CS$_2$; c) 2-mm cell of CB15; d) planar-aligned, 105-μm cell of 5CB with polarization of laser light parallel to the orientation of LC-molecules.

Figure 9. CCD-images for various samples in the focus.
From the images of Figure 9 (b-c) one can see the result of spatial self-phase modulation. For the sample in focus (and at the position corresponding to the maximum of the Z-scan curve) two effects are observed: 1) a high-contrast narrow diffraction ring outside the beam diameter; 2) a narrowing of the intensity envelope for isotropic LCs and CS2 of mm-scale pathlength. However, for the case of planar aligned nematic with polarization of laser light parallel to the orientation of the LC-molecules no such ring is observed at the routine Z-scan intensities (Figure 9, d). Owing to the larger values of β for parallel polarization, incident intensities in this case are kept at half of the values for the isotropic case (I = 0.36 GW/cm² for Figure 9,d). Further increasing of incident intensity above this level for the parallel polarization changes the far-field pattern significantly. From the Figure 10 one can see asymmetric scattering and elliptical ring formation. Major axes of this scattering (Figure 10 a,b) and the elliptical ring pattern (Figure 10,c) are perpendicular to the polarization of laser light and the orientation of LC molecules. The evolution of the pattern in time depends on the incident intensity, beam diameter, and, at given beam diameter, on the cell thickness. We observed the characteristic build-up time of the development of asymmetric scattering as ~ 0.3 – 1 sec for ~ 35 – 50-μm beam diameter by recording every pulse during this phase onto magnetic tape and digitizing individual frames of interest afterwards.

![Figure 10](image_url)

Figure 10. Far-field beam cross-sections with increasing incident intensity for the planar nematic 5CB cell (incident polarization parallel to the orientation of the LC-molecules). Polarization of laser light has vertical direction: a) I = 0.43 GW/cm², L = 25 μm; b) I = 0.48 GW/cm², L = 25 μm; c) I = 0.75 GW/cm², L = 105 μm.

At certain conditions, the divergence angle of the elliptical fringes abruptly increases, exceeding the spatial capture range of the camera’s detector. This necessitates capturing the fringe pattern on a white paper screen, and imaging the screen on to the detector by a conventional objective. Figure 10,c is the result of such imaging. For scale comparison, the absolute size of the initial beam spot observed clearly on all Figure 10 patterns does not change with the evolution of the pattern.

6. DISCUSSION AND CONCLUSION

This work showed that LCs may have a large value of transient nonlinear response even for several nanosecond timespan, but in the presence of nonlinear absorption this response depends both on the preparation procedure of the LC-cells, on the beam-waist diameter, and on the cell-thickness. For planar nematic LCs, it depends also on the laser-beam polarization, incident intensity and irradiation history (the time of measurement relative to onset of irradiation in multi-pulse exposures).
• The values for transient nonlinear refraction of both 5CB and chiral CB15 (same chemical composition) in their respective isotropic states, in the presence of nonlinear absorption, show the self-focusing-caused value (positive sign) of the nonlinear-refraction coefficient. For 5CB at 35.6 - 50°C the absolute value of n₂ is found to be 2.5 - 3.7 times larger than that of CS₂, and n₂ (CB15) = 2.5 n₂ (CS₂) at room temperature. For 6 - 9-ns pulse duration at 35 - 50-μm beam diameter, the orientational mechanism of nonlinearity dominates thermal and density-dependent ones. These results are different from the data of Reference 30 where, for much smaller beam waist radius, a negative value of nonlinearity was observed for isotropic 5CB. In this case thermal nonlinearity dominated because the smaller beam radius permitted temperature/density changes to develop within tₑ < t₀. Similar to current results, transient self-focusing for nanosecond pulse duration in the isotropic LC (MBBA) was also observed in References 19,22.

• For planar nematic 5CB with incident polarization parallel to the molecular orientation, we observed cumulative negative refractive nonlinearity at I ~ 0.36 GW/cm² with n₂ ~20 times higher than that for CS₂. At higher incident intensity, it increases its magnitude, and elliptical self-modulation rings evolve spatially. A large, negative value of nonlinear refraction for this case was observed also in References 30, 32-35. As to this nonlinearity, asymmetric scattering and elliptical ring formation, their strong polarization dependence (the effect exists only for parallel polarization) may be explained by the two-photon-absorption dichroism in the oriented layers of nematics (much stronger two-photon absorption for incident polarization parallel to the orientation of LC-molecules). In a speculative vein, in our paper 54, it was shown that cumulative heating of LC-material can take place. After more than tens of laser shots at 10-Hz repetition rate, the temperature of nematic LC, after stepwise incremental rises, attains a stationary value. This conclusion was made both from the experimental observation of stable spatial self-modulation rings pattern with build-up time (0.5-10 s), and numerical modeling solving the heat-transfer equation.

• Measurements of nonlinear absorption show the values of β of both 5CB (at 40 − 50°C) and CB15 (room temperature) in isotropic state to depend on the incident intensity and to vary from 4.5 - 30 cm/GW at I = 0.2 - 0.9 GW/cm² for mm-scale layer thickness. This variation of β (cm/GW) with the incident intensity is explained by the excited-state absorption mechanism. For picosecond excitation (28 ps), the value of β ~ 0.4 −0.8 cm/GW for I = 0.2 - 0.9 GW/cm² for planar nematic with incident polarization parallel to the molecular orientation. As to possible reason for larger values of β for nanosecond excitation, especially in the isotropic case, there are some speculations in the literature about triplet-state absorption which, for the nanosecond excitation, can increase the value of β in difference to the picosecond case. Unfortunately, existing spectroscopic data do not provide life-times and absorption cross-sections of levels of interest.

• Regarding possible reasons for the extraordinarily large β values (more than 100 cm/GW) observed first in Reference 30 and confirmed in this paper for “thin” (25 - 125-μm) layers of isotropic-state 5CB and CB15 (also planar-nematic 5CB), we believe that some additional energy dissipation mechanism develops in “thin” layers (surface effects), that is absent or has a smaller value in “thick” samples. For instance, scattering may contribute to the overall nonlinear changes in sample transmission. The precise mechanism of such scattering remains to be ascertained. However, one may speculate that local-heating-driven photodecomposition (carbonization) of the LC or small-scale (several μm) bubble formation 55-57 in the fluid itself, form the inhomogeneity that acts as scattering source. For instance, on rare occasions, we indeed observed near the cell surface ~0.5-mm-diameter bubbles in local coincidence with the area that the Z-scan beam had sampled. After surveying post-irradiation, “thin” cells under the microscope, we occasionally observed localized, carbonization sites of material. It seems more realistic that our “thick”-sample results a true manifestation of the nonlinear absorption of both isotropic 5CB and CB15 for several-nanosecond excitation. “Surface” effects is excluded for “thick” samples. The independence of β on layer thickness for 1-2-mm layers confirms this statement.

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