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Nanosecond Z-Scan Measurements of Optical Nonlinearities in 5CB and CB15 at 532 Nm

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Using the Z-scan method, we study the nonlinear absorption and refraction in the transient regime of 2 – 5-mm liquid-crystal samples in the isotropic state at 532 nm, in the *absence of linear absorption.* Simple model systems, pure 5CB and CB15, with identical chemical composition but conformal differences, were used. The 532-nm nonlinear refractive index of these compounds was found to be 2.4 – 3.7 times in excess of that of CS_2 for 7 – 8-ns pulse duration and 50 – 80-μm beam diameter. The nonlinear absorption coefficient of 5CB in its isotropic state is found to be more than 6 times larger for the 125-μm path length than for the 2-mm samples. Contributing mechanisms are discussed.

Keywords: z-scan; nematics; nonlinear absorption; refraction

1. INTRODUCTION

After initial experiments by Prost and Lalanne and Wong and Shen in the 1970-80's, a large number of experiments have been reported in more recent years on the nonlinear refraction and absorption of liquid crystals at nanosecond and picosecond timescales, both in the ordered and isotropic state, e.g., Ref.[1-4]. With regard to nonlinear absorption, it is by now a widely held notion, see, e.g., Ref.[2], that heating of pure liquid crystal materials, and various thermooptical effects connected with it, are driven at 532-nm by

two-photon absorption and concurrent or subsequent excited-state absorption. The conversion to thermal energy is explained by the efficient decay of excited states through radiationless-recombination channels. At the several-nanosecond timescale, molecular-reorientation (*transient*) and thermal and density refractive nonlinearities compete in changing the sign of the total refractive nonlinearity. For the different, given pulse durations, the influence of coupled thermal and density effects on the nonlinear refraction depends, through buildup time, on the interaction geometry[4], in particular the beam-waist diameter.

The purpose of this work is to extend previous experiments by the Kent State group[5-8] on highly efficient, laser-induced absorption by thin, nematic layers (both in the oriented and isotropic state) in the nanosecond regime to *longer-path-length* samples. In addition, there remain discrepancies between literature values for both the magnitude and the sign of the 532-nm, nonlinear refraction of isotropic-state 5CB and CB15 for nanosecond excitation that need to be remedied. These results are helpful for constructing intensity sensors for high-power lasers, see, e.g., Ref.[9].

2. EXPERIMENTAL

In the Z-scan method[10,11] 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. 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, this method, simple in principle, requires great care in ensuring reliable results[11], foremost in assuring verifiable beam quality and precise measurement of the beam caustic near focus[12]. 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.

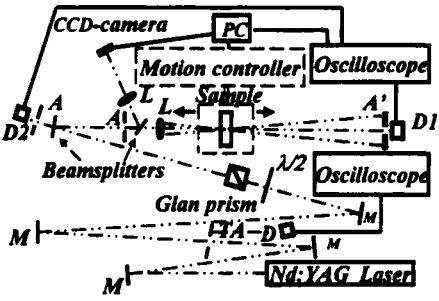

FIGURE 1 Experimental set up (mirrors M were used to increase path length to ~4 m).

The experimental set up used is shown in Figure 1. 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 and two, single-pass amplifiers, provided, at 10-Hz repetition rate, an output beam-spot diameter of ~8mm at 532nm. To optimize beam quality, a 1.5-mm-diameter Teflon aperture *TA* was placed in the central part of the beam[13], such that an Airy spot at a reasonable distance from the Teflon aperture was formed. A converging, 9.5-cm focal-length lens *L* was placed at 4 m from the Teflon

aperture. To remove side lobes, 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.

Signal *D1* and reference *D2* fast photodetectors (Hamamatsu S1722-01 and Motorola MRD510, respectively) or pyroelectric joulemeters (Gentec ED-100A with amplifier EDX-1, vendor recalibrated) were connected to a digitizing oscilloscope (Textronix 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 *D1*. Sometimes the entire photodetector was used as the aperture. For open-aperture measurements, we used an additional lens in front of *D1* 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 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 μm x 8.6 μm) was used in a separate, low-intensity, equivalent channel. Figure 2 shows a spatial distribution of the laser-beam intensity at the lens, beam spot size (diameter) ω versus the distance from the waist, and beam spot on the lens.

Samples were kept at elevated temperatures under control of an Instec heater control unit. It should be noted, that degassed 5CB served as sample material, having been filtered through a 5-μm filter in a clean room, prior to filling of the cell under regular laboratory conditions. Quartz cuvettes were used for "thick" samples and glass cells for "thin" ones.

3. RESULTS

3.1. "Thick" samples of 5CB in isotropic state

To exclude experimental errors we carried out these measurements for the same samples by two different sets of devices and data-acquisition methods, using the scheme of Figure 1.

(1) Results from Z-scan measurements of 2-mm 5CB cells at 45°C (phase transition temperature ~35.3°C[5]) are presented in Figure 3. These data resulted from pyroelectric joulemeters being used in both signal and reference channels. *D1* was placed at a distance of 65 cm from lens *L*, and for *A'* a 4.3-mm aperture was used. The transmittance factor S, see, e.g., Ref.[10] of this small-diameter aperture was ~0.1. For every translation-stage step (1 mm) 30 irradiation pulses were accumulated, and, after 5% windowing[11], these data were averaged. Dominated by software-run-time limits, a complete Z-scan experiment took 2 – 3 hours. We used the following parameters of incident laser radiation: 1) incident energy per pulse E = 13.5 μJ, pulse duration τ = 8 ns, minimal waist diameter at $1/e^2$ level, ω_o = 50 μm, incident peak intensity I_o = 0.34 GW/cm^2. For CS_2 E = 42 μJ, τ = 8 ns, ω_o = 50μm, I_o = 1.1 GW/cm^2.

(2) In difference with the above experiments, we also carried out Z-scan measurements with run times of only ~2.5 min. To shorten the experiment, the sample constantly moved and no real-time data averaging was performed. Upon completion of the entire run, results were filtered through 5% windowing. Fast photodiodes were used instead of joulemeters. For closed-aperture Z-scans, a 1-mm aperture was placed in front of the signal photodiode (4 mm detection area, 1-ns risetime), which was placed at the distance of 36 cm from the lens (S = 0.04). Incident energy was measured by a joulemeter. E = 55 μJ, τ = 7 ns, ω_o = 78μm, I_o = 0.66 GW/cm^2.

For deriving values of the nonlinear refraction coefficient γ and n_2 from the Z-scan curves, we used standard formulas from Reference[10] :

$$\Delta T = 0.406\, \Delta\Phi_o, \qquad (1)$$

where ΔT is the difference between the maximum and minimum values of the normalized transmittance T, $\Delta\Phi_o$ is the nonlinear phase shift on axis with the sample at the beam waist.

Here $\Delta\Phi_o = 2\pi\lambda^{-1}\gamma I_o L$, where λ is the wavelength, $I_o = 16E(\pi\omega_o^2\tau)^{-1}$ is the peak axial intensity at the waist, assuming a temporally and spatially Gaussian shaped pulse, L is the sample length, E is the pulse energy and τ is the pulse duration at FWHM.

The nonlinear absorption coefficient β was evaluated under the assumption that the two-photon absorption mechanism is the dominant driving mechanism, from the following formula[7-8]:

$$\Delta T_{abs} \cong -2^{-3/2}\beta I_o (1-R) L, \qquad (2)$$

where ΔT_{abs} is defined from the open aperture Z-scan curve, and R is the reflection coefficient.

As the accuracy of the Z-scan method critically depends on precise knowledge of the waist size, and precise calculation method should include beam profile, we carried out after each experiment, under identical experimental conditions, also Z-scan measurements for CS_2 whose well-known nonlinear refractive index $n_2 = 1.2.10^{-11}$ esu[10].

From data in Figure 3 we derive n_2 (5CB) = 3.2 n_2 (CS_2). For the experimental conditions of the second experiment, this ratio equals 3.7. In both cases, we observed self-focusing (converging lens). As to β, we resorted to expression (2) and found for the first case $\beta \cong 7.2$ cm/GW, and in the second case $\beta \cong 9.7$ cm/GW.

3.2. "Thick" samples of CB15 in isotropic state

The measurements on CB15 were carried out using constantly moving sample conditions (the time for each Z-scan was ~2.5 min, as in the case of the second experiment with 5CB), however, in both signal and reference channels the same joulemeters as in Figure 3 were used. We used 5-mm cuvette thickness. It should be noted, that for Z-scan measurements this thickness is not considered "thick", see, e.g. Ref.[11]. The calculations of Z-scan curves using formulas (1) and (2) yielded the following values: n_2 (CB15) = 2.4n_2 (CS_2), $\beta \cong 5.9$ cm/GW. E = 22.8 μJ, τ = 8 ns, ω_o = 65 μm, I_o= 0.34 GW/cm^2.

3.3. "Thin" layers of 5CB

As the above reported nonlinear-absorption-coefficients values for "thick" cells of isotropic 5CB are several times lower than had been obtained from thin layers in paper[5], we repeated measurements of Ref.[5] for thin layers of 5CB. We used planar aligned, 125-μm cells, using standard, rubbed polyimid coating for alignment agents. For the isotropic liquid crystal measurements we heated cell to the temperature 45°C.

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 polarization[5], orientation-dependent measurements were carried out.

Results from open-aperture Z-scans are presented in Figure 4:

a) for a planar-nematic 5CB layer irradiated by an incident beam polarization parallel to the director orientation (E = 14.7 μJ, τ = 7.5 ns, ω_o = 50 μm, I_o = 0.4 GW/cm^2) ;

b) after heating the cell of a) to the isotropic state (E = 41.4 μJ, τ = 7.5 ns, ω_o = 50 μm, I_o = 1.1 GW/cm^2). The experiments were made with the same devices and data acquisition as for Figure 3.

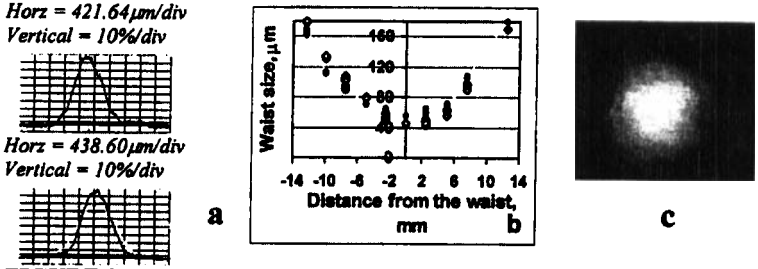

FIGURE 2 a) Beam profile at the lens L along X-axis (top) and Y-axis (bottom); b) Spot size (diameter) at $1/e^2$ level near the waist of the 9.5-cm focal-length lens (• - for X-axis of the beam, ◊ - for Y-axis); c) Cross-section of the beam spot at the lens.

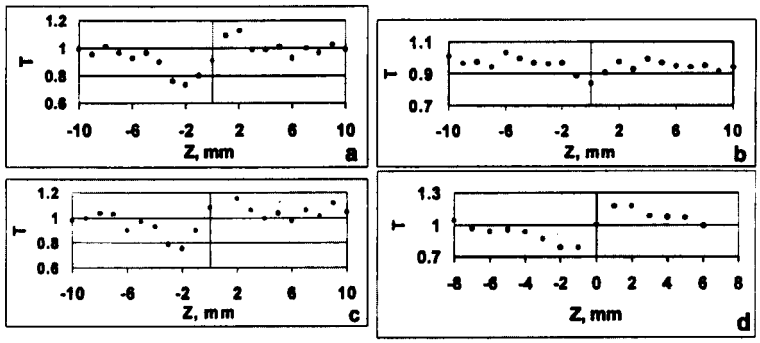

FIGURE 3 Z-scan curves for a 2-mm, isotropic (45° C), 5CB sample: a) closed aperture; b) open aperture; c) result of division of curve a) by curve b); d) Z-scan of CS_2 (2-mm cell).

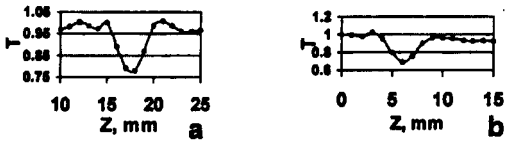

FIGURE 4 Open aperture Z-scan curves for the 125-μm layer of 5CB.

The results of calculations are as follows:

a) $\beta \cong 127.5$ cm/GW; b) $\beta \cong 60.2$ cm/GW. These data are in a good agreement with those of Ref.[5]. For the isotropic case, however, the value of β for the 125-μm cell was found to be more than 6 times larger than for the 2-mm cell in the current experiments. These differences *may not be*

explained by the absorption saturation effect for the "thick" samples. In the case of such saturation, an open-aperture curve should have a flat-bottom[11]. The calculated Rayleigh length of the focused beam $Z_0 = \pi\omega_0^2/4\lambda$ for ω_0 = 50 μm is equal to 3.7 mm, i.e., more than the 5CB-sample thickness (See also Fig. 2,b).

4. CONCLUSION

The values for transient nonlinear refraction of pure 5CB and CB15 in their respective isotropic states, in *the presence of nonlinear absorption*, offered by these Z-scan measurements show the *self-focusing*-caused value of the nonlinear-refraction coefficient to be 2.4 – 3.7 times larger than that of CS_2. For 7 – 8-ns pulse duration at 50 - 80-μm beam diameter, the orientational mechanism of nonlinearity prevails over the thermal and density ones. Transient *self-focusing* for nanosecond pulse duration in the isotropic liquid crystals (MBBA) was also observed in Ref.[14,15] for several-cm pathlengths.

Measurements of nonlinear absorption show the designer of optical-power-limiting, sensor- or other devices based on liquid crystals, that for nanosecond laser radiation the limiting effect may be increased if less than 0.1-mm (for 50 – 80-μm beam diameter) layers are used. Regarding possible reasons for the extraordinarily large nonlinear-absorption-coefficient values observed first in Ref.[5-8] and confirmed in our paper for "thin"-layer, isotropic-state 5CB, we believe, that some additional energy dissipation mechanism develops in "thin" layers that is absent or has 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 liquid crystal or small-scale (several μm) bubble formation[16] in the fluid itself, form the inhomogeneity that acts as scattering source. For instance, on rare occasion,

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.

As to the excited-state absorption mechanism which influence appears in the dependence of β (cm/GW) on the incident intensity[17] (for simplicity, results will not be presented here over an extended intensity range but will be published elsewhere), it is difficult to propose that a pulse-length increase from 28-ps[17] to several nanoseconds will entail a rise in β by more than 50 times when, for a layer thickness of ~ 0.1 mm, only two-photon absorption and excited-state absorption are taken into account. It seems more realistic that our "thick"-sample results which are in good agreement with Ref.[17], are, for ~GW/cm^2 incident intensities, a true manifestation of the nonlinear absorption of both isotropic 5CB and CB15.

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