Appl. Phys. B 78, 433–438 (2004)
DOI: 10.1007/s00340-003-1389-y

Applied Physics B
Lasers and Optics

Nonlinear refraction in CS_2

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Received: 2 May 2003/Revised version: 19 November 2003
Published online: 3 February 2004 •

ABSTRACT The nonlinear refractive index (γ) of CS_2 was measured using the Z-scan technique and laser radiation of various (femto-, pico-, and nano-second) pulse durations. We observed the growth of γ with the increase of the pulse duration (from $(3 \pm 0.6) \times 10^{-15}$ cm^2 W^{-1} at 110 fs to $(4 \pm 2) \times 10^{-14}$ cm^2 W^{-1} at 75 ns) due to the additional influence of the molecular reorientational Kerr effect in the case of longer (picosecond and nanosecond) pulses. Acoustic wave induced negative nonlinear refraction was observed using wavefront analysis. We analyzed the simultaneous influence of both electronic and molecular processes leading to the positive nonlinear refraction and acoustic processes leading to the negative nonlinear refraction in carbon disulfide. Variations of the refractive index due to the thermal effect at high pulse repetition rates were also investigated.

PACS 42.65.An; 42.65.Jx; 78.66.Nk

1 Introduction

For the past decade, the Z-scan technique has proved to be most convenient for nonlinear optical measurements due to its many attractive features (simplicity, high accuracy, low cost, well-elaborated theory, etc.). The opportunity to conduct simultaneous measurements of various nonlinear optical parameters in one set of experiments also makes this technique attractive and applicable for different materials. An important parameter in these measurements is the laser pulse duration. The measurements performed using different laser pulse durations have shown the different values of nonlinear optical parameters, in particular, the nonlinear refractive index (γ). From here, the problem of determining the role of this parameter in such measurements arises.

Meanwhile, carbon disulfide has long been considered to be a reference material for the measurements of nonlinear refractive indices. Repeated measurements of this parameter in CS_2 at the initial stages of nonlinear optical studies have shown that its γ value varied in the range of units of 10^{-14} cm^2 W^{-1} (see [1] and references therein). These studies were carried out using pico- and nanosecond pulses. However, the last measurements using femtosecond radiation have shown a decrease of this value for more than one order [2–4]. At the same time, even in this temporal range (~ 400 fs), some studies have shown considerably different results (from 1.3×10^{-16} cm^2 W^{-1} [5], to 12×10^{-15} cm^2 W^{-1} [6], and further 4×10^{-14} cm^2 W^{-1} [7]).

In this paper, we report on our studies of γ of CS_2 using laser pulses of different pulse duration (femtosecond, picosecond, and nanosecond) in one set of Z-scan measurements. The mutual influence of self-focusing and self-defocusing processes will be discussed in the case of 8-ns pulses. We show the simultaneous appearance of both electronic and molecular processes leading to the positive nonlinear refraction and acoustic processes leading to the negative nonlinear refraction. We also demonstrate the influence of thermal lens caused by two-photon absorption on the γ sign at high pulse repetition rates.

2 Experimental arrangement

We used two lasers operating at different lasing conditions. The first one (L1) was the hybrid Nd : glass–Ti : sapphire laser (L1) (seeding oscillator, GLX-200, TimeBandwidth + CPA Ti:sapphire regenerative amplifier) generating 475-fs, 1054-nm, 7-μJ pulses at 1-Hz repetition rate. Uncompressed radiation from this laser ($t = 300$ ps, $E = 10$ μJ) was used for investigation of CS_2 nonlinearities in picosecond range. We also used 100-MHz, 280-fs, 100-mW, 1054-nm radiation from a seeding oscillator (GLX-200) for investigation of the thermal effect induced nonlinear refraction in CS_2. Another laser (L2) was the Ti : sapphire laser (seeding oscillator, Tsunami + CPA TSA-10F regenerative amplifier with double passed linear amplifier, Spectra Physics) delivering femtosecond pulses ($t = 110$ fs, $\lambda = 795$ nm, $E = 10$ mJ) with 10-Hz pulse repetition rate. The output radiation with variable pulse duration (from 110 fs to 1.6 ps) was also available from this laser. The laser was also operated at Q-switched regime delivering both 8-ns (10 mJ) and 75-ns (0.25 mJ) pulses. 80-MHz, 100-fs, 300-mW pulses were available from the seeding oscillator (Tsunami) for investigation of thermal-induced self-interaction in CS_2.

The conventional Z-scan scheme was used in these studies. Laser radiation (either at the wavelength of 795 nm or

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1054 nm) was focused by a 20-cm focal length lens. Investigated samples (CS_2-filled silica glass cells) were moved by a translating stage along the Z-axis. The energy of laser pulses was measured by a calibrated photodiode. A 1-mm aperture was kept at a distance of 70 cm from the focal plane (closed-aperture Z-scheme). The radiation propagated through this aperture was registered by a second photodiode. The radiation energy registered by the second photodiode was normalized with respect to the radiation energy registered by the first photodiode, to avoid the influence of non-stability of the laser parameters. The closed-aperture Z-scan scheme allowed the determination of both the sign and the value of γ of carbon disulfide.

3 Results

The first set of experiments was carried out using laser L1. Closed-aperture Z-scans (Fig. 1) have shown the self-focusing features of carbon disulfide in case of 1054-nm, 475-fs pulses. The influence of nonlinear absorption was clearly seen in these experiments at intensities of $I_o \geqslant 1.7 \times 10^{10}\,\mathrm{W\,cm^{-2}}$ (curve 3).

One of the advantages of the Z-scan technique is the possibility of distinguishing nonlinear refraction and nonlinear absorption. In general, the normalized transmittance dependence can be presented as follows [8]:

$$T = 1 + \frac{2(-\varrho x^2 + 2x - 3\varrho)}{(x^2+9)(x^2+1)}\Delta\Phi_o \qquad (1)$$

where T is the normalized transmittance of the sample, $x = z/z_o$, $\varrho = \beta/2k\gamma$. k and z_o are the wave number, $k = 2\pi/\lambda$, and the diffraction length of the beam, $z_o = 0.5\,k\,\omega_o^2$, respectively. Here ω_o is the beam waist radius (at $1/e^2$ intensity level). β (nonlinear absorption coefficient) and γ related with the phase changes due to nonlinear absorption ($\Delta\psi_o$) and nonlinear refraction ($\Delta\Phi_o$) through the relations $\Delta\psi_o = \beta I_o L_{eff}/2$ and $\Delta\Phi_o = k\gamma I_o L_{eff}$. Here I_o is the laser radiation intensity in the focal plane, $L_{eff} = [1 - \exp(-\alpha_o L)]/\alpha_o$ is the effective length of the sample, α_o is the linear absorption coefficient, L is the sample length (in our case $L = 2$ mm). This relation was used for fitting with experimental data.

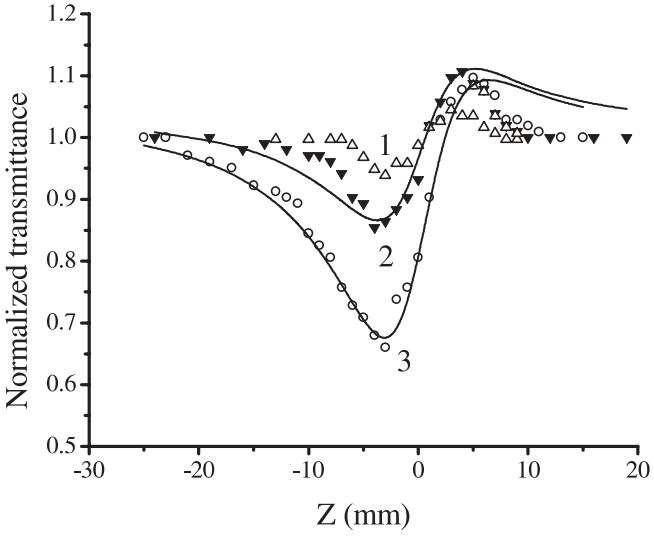

FIGURE 1 Normalized transmittance dependences (L1, $t = 475$ fs) of CS_2 using the closed-aperture Z-scheme at different intensities. 1 – $7 \times 10^9\,\mathrm{W\,cm^{-2}}$, 2 – $1.4 \times 10^{10}\,\mathrm{W\,cm^{-2}}$, 3 – $3 \times 10^{10}\,\mathrm{W\,cm^{-2}}$

γ and β were calculated to be $(3.5 \pm 0.7) \times 10^{-15}\,\mathrm{cm^2\,W^{-1}}$ and $8 \times 10^{-14}\,\mathrm{cm\,W^{-1}}$, respectively. The latter parameter was measured at $I_o = 3 \times 10^{10}\,\mathrm{W\,cm^{-2}}$. Our studies have shown a negligible dependence of γ on laser intensity, whereas the β measurements indicated a strong dependence on the intensity used, pointing out the influence of high-order nonlinear processes (other than two-photon absorption). The separate measurements of nonlinear absorption at this wavelength (1054 nm) were carried out using an open-aperture Z-scheme. These studies have confirmed that the three-photon absorption process was responsible for nonlinear absorption ($\beta^{(3\omega)} = (2.8 \pm 1.4) \times 10^{-23}\,\mathrm{cm^3\,W^{-2}}$).

The investigations of nonlinear refraction using uncompressed 300-ps pulses from laser L1 have again shown the self-focusing properties of CS_2. These measurements were carried out close to the threshold of registration of $T(z)$ variations due to small intensity of laser radiation. The γ in that case was measured to be $(3.2 \pm 1.6) \times 10^{-14}\,\mathrm{cm^2\,W^{-1}}$, which was almost one order higher than that measured using femtosecond pulses.

We tried observing the thermal lens induced self-defocusing in CS_2 using pulses of 100-MHz repetition rate ($t = 280$ fs, $\lambda = 1054$ nm, $W = 100$ mW) from laser L1 seeding oscillator (GLX-200, TimeBandwidth). However, in that case the peak intensity of single pulse was too small ($10^9\,\mathrm{W\,cm^{-2}}$) to induce the three-photon absorption and the thermal lens appearance in this medium.

The next set of experiments was carried out using laser L2, operated in Q-switched regime ($\lambda = 795$ nm, $t = 8$ ns). Note that all our measurements of nonlinear refraction were carried out in conditions of "thin" medium, which means the fulfillment of the relation $L < z_o$ [9]. The change of L2 lasing regime from femtosecond to nanosecond pulses generation led to the decrease of both output divergence and diffraction length of the beam (from $z_o = 2.6$ mm, for femtosecond pulses, to $z_o = 1.8$ mm, for nanosecond pulses). In the case of nanosecond pulses, the $r = L/z_o$ ratio became equal to 1.1. The analysis of the usefulness of Z-scan relations (1) in conditions of "thick" medium was carried out in [10, 11]. It was shown that the peak and valley distance in Z-scans changes from $\Delta Z_{p\text{-}v} \approx 1.7 z_o$ for small r to $\Delta Z_{p\text{-}v} \approx r z_o = L$ for large r. Our calculations have shown that at $r = 1.1$ this distance changes insignificantly (from $1.7 z_o$ to $1.87 z_o$). It means that in our case, we can use the same relations of the standard Z-scan theory for analysis of nonlinear refraction caused by nanosecond pulses.

Figure 2 presents the closed-aperture Z-scans of CS_2 using 8-ns pulses. These measurements were carried out at intensities up to $I_o = 1.5 \times 10^{10}\,\mathrm{W\,cm^{-2}}$. The surface optical breakdown was observed at higher intensities. The small asymmetry of the Z-scans was caused by a nonlinear absorption (Fig. 2, curve 2).

The value of γ in that case was calculated to be $(3.5 \pm 0.7) \times 10^{-14}\,\mathrm{cm^2\,W^{-1}}$ from the theoretical fits of several Z-scans (Fig. 2) measured at different intensities of 8-ns pulses. This value was again one order higher than that in the case of femtosecond pulses. The separate measurements of sil-

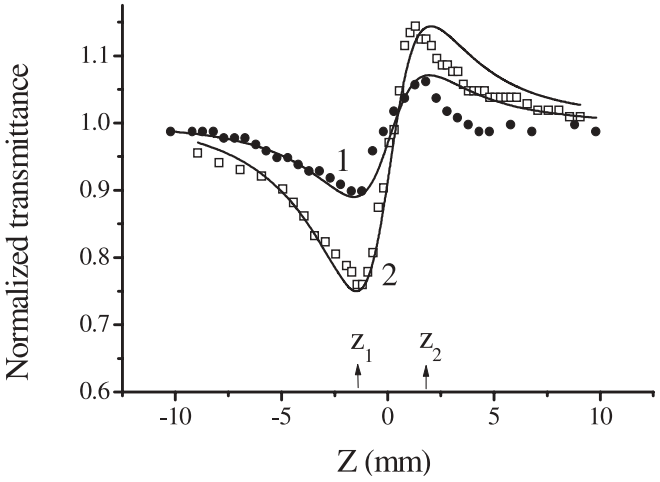

FIGURE 2 Normalized transmittance dependences (L2, $t = 8$ ns) using the closed-aperture Z-scheme at different intensities. $1 - 8 \times 10^8$ W cm^{-2}, $2 - 1.7 \times 10^9$ W cm^{-2}

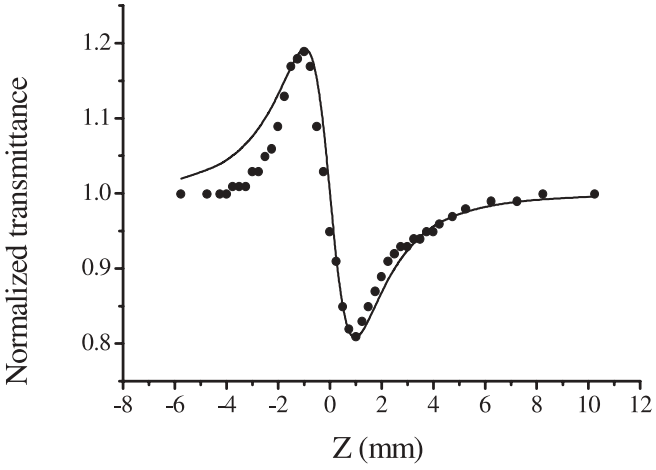

FIGURE 3 Normalized transmittance dependence using high repetition rate pulses (L2, $t = 100$ fs, 80-MHz)

ica glass (without CS_2) were carried out using nanosecond radiation and no nonlinear refraction in this medium was registered at intensities up to 1×10^{10} W cm^{-2}. 75-ns pulses were also available from laser L2. The energy of these pulses (2.5×10^{-4} J) was considerably smaller than that in the case of 8-ns pulses. The γ in that case was measured to be $(4 \pm 2) \times 10^{-14}$ cm^2 W^{-1}.

The previously observed influence of thermal lens on nonlinear optical refraction in CS_2 at high repetition rates of femtosecond pulses (of order of tens MHz [2, 4]) have shown the importance of acoustic waves generation and heat accumulation due to various (linear and/or nonlinear) types of absorption. We carried out the Z-scan studies using radiation from the laser L2 seeding oscillator (Tsunami) operated at 80-MHz pulse repetition rate. Figure 3 presents the closed-aperture Z-scan using 100-fs, 80-MHz, 260-mW, 795-nm radiation indicating the self-defocusing of such radiation in carbon disulfide ($\gamma = -(1 \pm 0.2) \times 10^{-14}$ cm^2 W^{-1}).

The γ variations at different intensities were analyzed using laser L2 (10-Hz pulse repetition rate, 110-fs 795-nm). These measurements were carried out in a broad intensity range of 1×10^9 W cm^{-2} to 1×10^{12} W cm^{-2} (Fig. 4). The only nonlinear process observed at small intensities was the nonlinear refraction. A difference between the normalized peak and valley transmittance ($\Delta T_{\text{p-v}}$) linearly depended on laser intensity. However, at $I_o > 1 \times 10^{10}$ W cm^{-2} the nonlinear absorption affected the whole picture of normalized transmittance (Fig. 4, curve 3). These measurements were carried out up to the intensities when the saturation of nonlinear refraction became significant (Fig. 4, curves 4 and 5).

The solid lines corresponding to the intensities at which the saturation processes were insignificant (Fig. 4, curves 2 and 3) are the theoretical fits calculated for the medium with third-order nonlinearity. The values of γ and β were calculated to be $(3 \pm 0.6) \times 10^{-15}$ cm^2 W^{-1} and $(4.2 \pm 1.3) \times 10^{-11}$ cm W^{-1}, respectively. The intensity dependent measurements of γ have shown the independence of this parameter on laser intensity till $I_o = 1 \times 10^{11}$ W cm^{-2}. At higher intensities, the decrease of the nonlinear refractive index with intensity growth was observed, probably due to influence of high-order processes.

Attention was given to the investigation of the contribution from silica glass cell; however, up to the intensities of $I_o = 4 \times 10^{10}$ W cm^{-2} we did not observe any significant variation

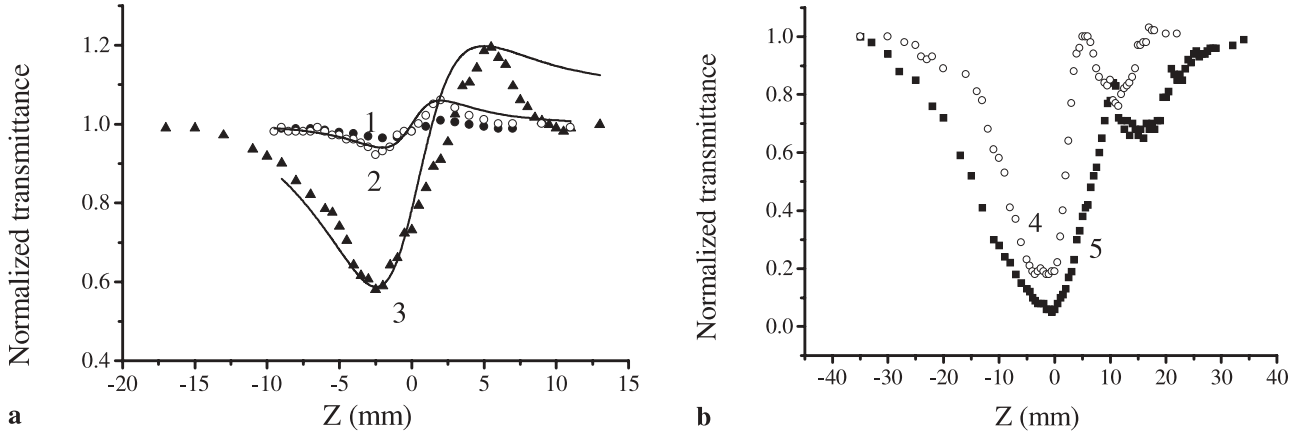

FIGURE 4 Normalized transmittance dependences (L2, $t = 110$ fs) using the closed-aperture Z-scheme at different intensities. $1 - 2.6 \times 10^9$ W cm^{-2}, $2 - 8 \times 10^9$ W cm^{-2}, $3 - 3.5 \times 10^{10}$ W cm^{-2}, $4 - 3 \times 10^{11}$ W cm^{-2}, $5 - 9 \times 10^{11}$ W cm^{-2}

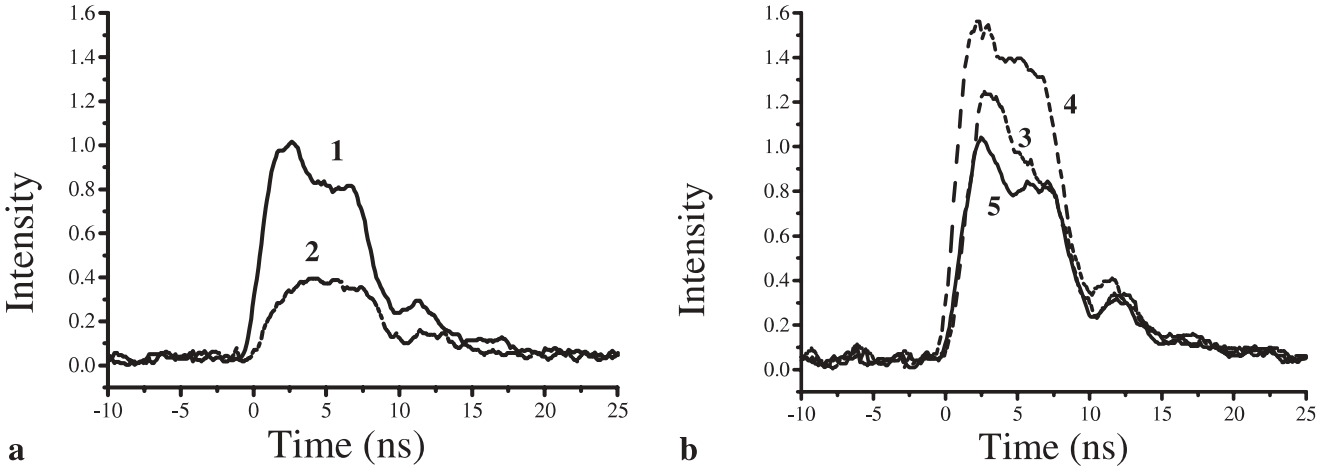

FIGURE 5 Temporal waveforms of laser pulse transmitted through an aperture at different positions of CS_2–filled cell. The description is given in the text

in normalized transmission measurements of such medium without carbon disulfide. Our separate measurements of silica glass nonlinearity have shown that γ of this medium was $(2 \pm 0.4) \times 10^{-16}\,\mathrm{cm^2\,W^{-1}}$.

We varied the pulse duration of laser L2 by changing the distance between gratings in the compressor. Our measurements at two different pulse durations in the femtosecond range (200 fs and 600 fs) did not show any variations of γ within the accuracy of the measurements of this parameter (20%). At the same time, our measurements using 1.6-ps pulses have shown a visible growth of nonlinear refractive index ($\gamma = (1.2 \pm 0.24) \times 10^{-14}\,\mathrm{cm^2\,W^{-1}}$).

4 Discussion and conclusions

There are some fast processes (intermolecular interaction, molecular reorientational Kerr effect, electronic Kerr effect) that can originate from the nonlinear optical response of CS_2 molecules and contribute to the overall nonlinear adding of the refractive index. The slowest among them is the process connected with reorientation of molecules. Previously, the relaxation times of these three processes were reported as 1.5 ps for reorientation of molecules [12], and ~ 200 fs for other processes [6]. Our results indicating the increase of γ at a pulse duration of $t = 1.6$ ps have confirmed these estimations of the addition of fast nonlinearities with the growth of pulse duration above the femtosecond range.

The unfixed value of the nonlinear refractive index in our CS_2 studies at high intensities has shown that the origin of nonlinear refraction in such conditions should not be attributed only to third-order nonlinearity, but also to higher orders of nonlinearity induced by the intermolecular and intramolecular interaction as well as by the appearance of excited states and free carriers generated by high intense radiation.

Among a variety of the above-mentioned optical processes contributed at short laser pulses to the nonlinear refraction of CS_2 the Kerr-induced effect, which is due to the electronic response of molecules, seems to be most important. Considerable nonlinear contribution to the refractive index (n) at appropriate experimental conditions can also be due to the thermal effect. This effect consists of two parts: one is the "fast" process of acoustic wave propagation, and other is the "slow" steady state changing of medium density due to accumulative thermal heating of the absorbing area. The slow process should be taken into consideration at low thermal conductivity and/or high pulse repetition rates. The fast process causes the matter density variations due to acoustic wave propagation after local heating. The rise time (τ), which is necessary for observing the density variations and further the refractive index variations, is determined by a ratio of beam waist radius at the focal point to the sound velocity in the medium ($V_s : \tau = \omega_o / V_s$). Taking into account our experimental conditions ($\omega_o = 21$ μm for 8-ns pulses at the wavelength of 795 nm, $V_s \sim 1500$ m/s), a rise time can be estimated as $\tau \sim 14$ ns, i.e., the influence of the thermal effect should not be neglected in the case of nanosecond pulses. Previous studies of this phenomenon in CS_2 using long nanosecond pulses (200 ns, [9]) have shown the influence of thermal nonlinearity resulting from linear absorption in this medium in the far IR range ($\alpha = 0.22\,\mathrm{cm^{-1}}$, $\lambda = 10.6$ μm). In our case, the influence of linear absorption is insignificant. Our spectral measurements have shown that the linear absorption coefficient of CS_2 at 795 nm ($\alpha = 0.006\,\mathrm{cm^{-1}}$) is too small to cause the thermal lens appearance. However, there is another option in the growth of absorption, a nonlinear absorption.

The technique previously proposed to reveal a slow nonlinear mechanism in Z-scan-like γ measurements [13, 14] was based on the analysis of the temporal waveform of the beam transmitted through an aperture in the closed-aperture Z-scan scheme. Part of the radiation self-focuses (or self-defocuses) inside the cell when the energy density reaches its definite value at the appropriate point of the Z scale. As a result, the smaller (or bigger) part of the whole radiation passes through the aperture and registers by a fast registrar. Thus, the type of nonlinearity can be estimated by analyzing the variations of temporal shape of propagated pulse when the sample placed at the peak or valley position of the Z-scan. Previously this technique allowed for the distinguishing of thermal nonlinearities for methyl nitroaniline [13] and colloidal metal nanoparticles [14]. We used analogous technique for the detection of the thermal effect induced nonlinearity using 8- and 75-ns pulses ($\lambda = 795$ nm). The variations of the temporal form of exciting pulses in the case of 8-ns pulses were observed. We were

able to register the mutual influence of both positive refraction (Kerr-induced variations of n) and initial stages of negative refraction (acoustic wave induced variations of n).

Figure 5.1 presents the temporal trace of 8-ns pulse (LeCroy 9362, 10 Gs/s) in the case when a sample was located far from the focal area. With increasing proximity to the focal plane, the drop of the overall intensity of the pulse occurred indicating the positive nonlinear refraction. Just after the sample moved through the position corresponding to T_v (z_1, see Fig. 2, minimum transmittance at a valley of Z-scan), the trailing part of temporal shape increased with respect to the leading part (Fig. 5.2). This increase is a manifestation of nonlinear absorption starting to play an important role in overall propagation of radiation through the aperture placed before the fast photodiode. An acoustic wave generated by nonlinear absorption spreads through the beam waist and decreases the refractive index ($\gamma < 0$). The overall process seemed to be a positive nonlinear refraction dominated by fast Kerr-induced processes due to insignificance of nonlinear absorption and the thermal lens generation at intensities used. Next, with further movement of the sample and before reaching the position corresponding to T_p (z_2, maximum transmittance at a peak of Z-scan), the reverse reshaping of the trailing part of the temporal waveform have appeared (Fig. 5.3). At $z = z_2$ this pulse re-shaping became insignificant (Fig. 5.4) and then fully disappeared with further movement of the sample far from z_2 (Fig. 5.5), together with the influence of fast electronic and reorientational processes.

All these variations of waveform indicate the manifestation of transient negative nonlinear refraction induced by acoustic wave. Taking into account the independence of measured $\Delta T_{p\text{-}v}$ value on laser radiation fluence, we can conclude that nonlinear absorption induced variations of the refractive index did not sufficiently contribute to the overall "effective" nonlinear refractive index with respect to fast nonlinear optical processes. No changes in waveform shape of the pulse transmitted through the aperture in closed-aperture scheme were observed at intensity of twice below the used one (4×10^9 W cm^{-2}). These studies have shown that acoustic wave induced variations of the refractive index became visible ~ 5–7 ns after the beginning of propagation through the medium that is two times smaller than the estimated time of the acoustic wave propagation (14 ns). Our observations have shown the growing influence of this process with intensity increasing through the pulse. This peculiarity as well as the distance between the maximum and minimum positions of trailing part variations, which appeared to be shorter than $1.7z_o$, confirm the assumption of the involvement of two-photon absorption process in acoustic wave induced negative nonlinear refraction in CS_2. No analogous features were observed using 75-ns pulses due to their small intensity. The two-photon absorption of 795-nm radiation in CS_2 seems to be a reasonable assumption taking into account the coincidence of two-fold frequency ($\lambda = 397$ nm) of excited radiation with the broad absorbance band of carbon disulfide in UV range ($290 \div 410$ nm [15]).

All above presented Z-scans (with the exception of Fig. 3) were carried out using the laser pulses operated at repetition rates of 1 Hz and 10 Hz. We performed similar scans with single shots and no difference was found indicating that long-term phenomena (due to thermal accumulation) were negligible under our experimental conditions.

We attribute the negative sign of γ in experiments with 80-MHz pulse repetition rate to the thermal lensing effect, as was also confirmed in previous studies. As it was mentioned above, the appearance of the thermal lens can be due to both acoustic wave induced density variations and thermal accumulation effects caused by heat accumulation between pulses. The CS_2 acoustic transit time in our experimental conditions was of order of 14 ns. This means that the density variations at 80-MHz pulse repetition rate (the distance between pulses of $\Delta t = 12.5$ ns) were involved in the change of refraction. The difference between the acoustic wave induced mechanisms of density changes in the cases of 8-ns low-repetition rate pulses and 100-fs high-repetition rate pulses is that in the former case the nonlinear optical process occurs due to self-interaction of the pulse propagated through the medium, whereas in latter case the pulse interacts with the acoustic wave generated by the previous pulse propagated through the medium 12.5 ns before the pulse of consideration.

The time interval of 14 ns means the time of acoustic wave propagation through the whole beam waist area. This time interval is longer then the distance between laser pulses at 80-MHz pulse repetition rate. However, there is no necessity of density variations inside the whole volume of beam waist to observe the influence of variations of the refractive index. Even in the case when the acoustic wave varies, the density of 1/3 part of the beam waist (it can be done for a period of ~ 5 ns), these density variations in the central part of the beam waist induce the observable variations of the refractive index. These assumptions have been confirmed in previous studies of colloidal metals [14]. The confirmation of this conclusion can also be found in the waveform traces presented in Fig. 5. One can see that thermal induced waveform variations have started to play an important role 5 ns after the beginning of pulse propagation through the medium (compare the shapes of oscilloscope traces 1 and 2, as well as 5 and 3).

Another process involved in this case is the thermal accumulation due to nonlinear absorption. The time required for thermal conduction effects to become significant is defined as [2] $\tau = \omega_o^2 \varrho' c_p / 4\kappa$, where c_p is the specific heat, ϱ' is the density, and κ is thermal conductivity. In our experimental conditions ($\omega_o = 21$ μm, $\varrho' = 1.263$ g cm^{-3}, $c_p = 0.995$ J g^{-1} K^{-1}, $\kappa = 0.16$ W m^{-1} K^{-1}) $\tau \approx 1$ ms, thus indicating the importance of the accumulative thermal processes in the case of 80-MHz pulse repetition rate regime of laser-matter interaction.

We are inclined to conclude about the prevailing role of acoustic wave induced variations of n rather than of accumulative processes induced changes of the refractive index. This conclusion is drawn on both the discrepancy between our dn/dT calculations (-1.2×10^{-5} K^{-1}) based on the assumption of the leading role of the accumulative processes in variations of n and previous measurements of this parameter based on steady state thermodynamic conditions (-8.3×10^{-4} K^{-1} [9]). The ratio of these parameters (-1.2×10^{-5} K^{-1} / -8.3×10^{-4} K^{-1} ≈ 0.015) can serve as a rough estimation of the influence of "slow" accumulative processes with respect to "fast" acoustic wave induced pro-

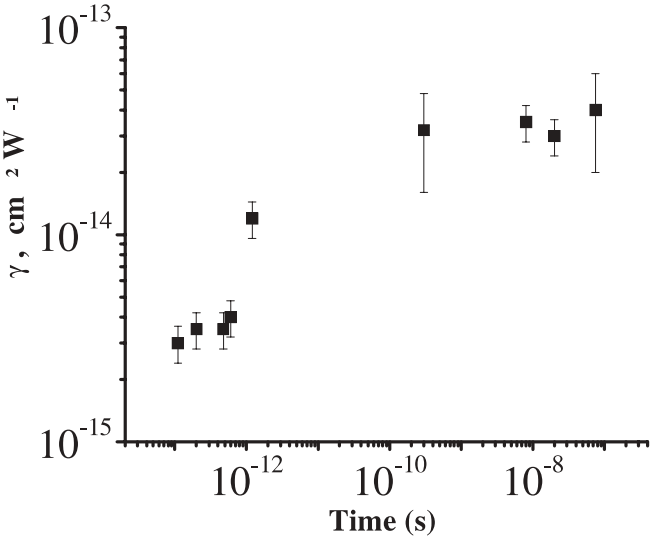

FIGURE 6 The dependence of CS_2 nonlinear refractive index on pulse duration

cesses in the overall negative nonlinear index of CS_2 caused by thermal effects.

The results of our measurements of the nonlinear refractive index of CS_2 using radiation of different pulse duration are summarized in Fig. 6. Here we also included the results of our γ measurements using second harmonic of Nd : YAG laser radiation ($\lambda = 532$ nm, $t = 20$ ns, $\gamma = 3 \times 10^{-14}$ cm^2 W^{-1}). Note the insignificant spectral dispersion of γ in visible (532 nm) and near IR (795 nm and 1054 nm) ranges. We excluded our results indicating the influence of slow nonlinearities caused by the thermal effect, thus limiting our comparison to the influence of fast nonlinearities measurements (molecular reorientational Kerr effect, electronic effect, and intermolecular interaction). These fast processes were previously considered as the main origins of the optical nonlinearity [12]. Our measurements have shown that in femtosecond time range, the contribution of molecular reorientation becomes insignificant due to its relaxation time exceeding 1.5 ps. In the case of picosecond and nanosecond pulses, this effect contributes to overall nonlinear refractive index.

In conclusion, we investigated the nonlinear refractive index of CS_2 using the Z-scan technique and laser radiation in various (femto-, pico-, and nanosecond) pulse durations. We observed the growth of γ with increase of the pulse duration (from $(3 \pm 0.6) \times 10^{-15}$ cm^2 W^{-1} at 110 fs to $(4 \pm 2) \times 10^{-14}$ cm^2 W^{-1} at 75 ns) due to the additional influence of the molecular reorientational Kerr effect in case of the longer (picosecond and nanosecond) pulses. Acoustic wave induced negative nonlinear refraction was analyzed using pulse waveform variations in the closed-aperture Z-scheme. We observed simultaneous influence of both electronic and molecular processes leading to the positive nonlinear refraction and acoustic processes leading to the negative nonlinear refraction. The variations of refractive index due to the thermal effect at high pulse repetition rates were also investigated.

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