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High Repetition-Rate Laser Z-Scan Measurements: Criterion for Recognizing Adverse Thermal Effects

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Abstract

This study reports z-scan measurements in which high-repetition-rate and reduced repetition-rate (pulse-picked) laser beams were employed using a standard sample, carbon disulfide (CS_2) contained in supposedly high damage-threshold spectroscopic-grade pristine fused-quartz cuvettes. The results suggest that at reduced repetition rates, the closed aperture z-scan profile for CS_2 displays the expected configuration. However, at high repetition-rates the closed aperture z-scans are distinctly different resulting in unexpectedly large nonlinear refractive index of CS_2 due to thermal effects that resulted in below-threshold laser-induced damage of the cuvettes. Normaski microscope images confirm the damage and open aperture z-scan study of the damaged fused-quartz cuvettes yielded about two orders of magnitude enhancement in the nonlinear absorption coefficient of silica. Based on these findings, the necessary criterion for recognizing below-threshold laser-induced damage in any high-repetition-rate laser z-scan measurement has been formulated in order to help avoid erroneous interpretation of the origin and strength of nonlinear response in such studies.

Keywords: Z-Scan, Damage-Threshold, Laser-Induced Damage, Nonlinear Refractive Index, Nonlinear Absorption Coefficient, Repetition-Rate

1. Introduction

The search for materials that have optical limiting properties for possible optoelectronic and optical limiting applications has recently become an active field of research in which several methods have been employed for studying the third-order optical susceptibility of such materials (Jafari, Zeynizadeh, & Darvishi, 2018; Smirnova, Rudenko, & Hryn, 2017; Dissanayake, Cifuentes, & Humphrey, 2018; Reyna et al., 2018; Gao & Kong, 2018). Some of these techniques are fairly sensitive but require complex experimental setups (Solati, Savadkoohi, & Dorranian, 2018; Vincent, Petit, & Chin, 2002; Samoc et al., 1998) while the others are relatively insensitive and entail detailed wave propagation analysis (Sheik-Bahae, Said, Wei, Hagan, & Van Stryland, 1990). In view of this, the nonlinear optical characterization technique of choice is usually the z-scan (Falconieri & Salvetti, 1999); a simple and sensitive single-beam method that uses the principle of spatial beam distortion to measure both the real and imaginary parts of complex indices of refraction and their signs. This approach requires tight-focusing of the probe beam into the test material in order to induce the expected nonlinear response. Both solid and liquid samples are suitable candidates for such studies with the liquid usually kept in cuvettes of appropriate laser-induced damage (LID) thresholds.

It is known that the use of lasers of high pulse repetition-rate frequencies in determining nonlinear optical properties of materials gives rise to cumulative effects capable of completely modifying the nonlinear behavior of test samples (Gnoli, Razzari, & Righini, 2005; Ganeev et al., 2004). At these frequencies, the properties of the transmitted light are determined by the linear and nonlinear response of the medium, and possible thermo-optical effects caused by even very low single and multiple-photon absorption of the laser energy (Genin, Feit, Kozlowski, Rubenchik, Salleo, & Yoshiyama, 2000). This work reports on the study of the influence of high laser repetition-rate on nonlinear response of test materials as well as apparently high damage threshold fused-quartz cuvettes in which samples are contained during z-scan measurements. Furthermore, the study presents images of the damage morphologies that suggest that that under certain experimental conditions, some of the supposedly high LID materials do experience below-threshold damage.

2. Z-Scan Theory

Consider a nonlinear material of intensity-dependent refractive index n expressed as:

$$\begin{aligned} n &= n_0 + n_2 < E^2 > \\ &= n_o + \gamma I \end{aligned} \quad (1)$$

where n_o is the linear index of refraction, n_2 is the nonlinear component, $<E^2>$ is the time-averaged square of the electric field of the beam (of intensity I) expressed in electrostatic units (esu). The change in the index of refraction caused by the presence of the high intense beams forms the basis of an intensity-dependent phenomenon known as self-focusing. Such an intense beam of light propagating in a material medium modifies the optical properties of the medium in such a manner that the beam is caused to come to a focus in the material. This requires $n_2 > 0$ and the material medium basically behaves as a convex lens. The reverse effect, self-defocusing, occurs in $n_2 < 0$ materials. In general, the physical origin of nonlinear refraction can be electronic, molecular, electrostrictive or thermal. Thermal nonlinearities are commonly slow and thus much larger than the electronic nonlinearities of the same material, often by several orders of magnitude. However, thermal time-constants are of magnitude longer than those of their electronic counterparts making them less useful for many high speed optoelectronic applications.

The on-axis nonlinear phase shift experienced by the incident beam at the focus is expressed as

$$\Delta\Phi_0(t) = k\Delta n_0(t) L_{\text{eff}} \quad (2)$$

where $L_{\text{eff}} = (1 - e^{-\alpha L})/\alpha$ represents the effective length of the sample, $\Delta n_0 = \gamma I_0(t)$ with $I_0(t)$ being the on-axis intensity at the focus (z = 0), k =2π/λ, and L is the length of the nonlinear sample of linear absorption coefficient α.

There are two types of z-scan measurement: closed aperture (CA) and open aperture (OA) z-scans. In a CA z-scan, an aperture is positioned in the far-field and the power transmitted through the nonlinear medium is measured as a function of the sample's position (z), with respect to the focal plane. This arrangement is used in measuring the sign and magnitude of the nonlinear refraction (n_2). Figure 1 shows a schematic of a closed aperture z-scan experimental setup.

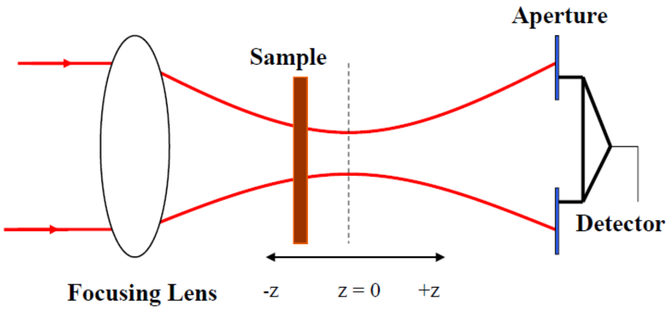

Figure 1. A schematic of closed aperture z-scan experimental setup

For small phase distortions ($|\Delta\Phi_o| << 1$) and small aperture (S≈0), the extrema (peak and valley) of the on-axis CA z-scan transmittance (T_{p-v}) can be calculated by solving the transmittance equation, $dT(z, \Delta\Phi_o)/dz = 0$. The result is expressed by the relation

$$\Delta T_{p-v} = 0.406|\Delta\Phi_0| \quad (3)$$

To the first order in intensity, the transmitted intensity can be expressed as

$$T(z, \Delta\Phi_0) = 1 + \frac{4x\Delta\Phi_0}{(1+x^2)(9+x^2)} \quad (4)$$

where $x = z/z_0$ with z_0 being the Rayleigh range.
For OA z-scan, the aperture is removed and the transmitted intensity is no longer sensitive to the beam distortion and the data is a function of nonlinear absorption. The effect of nonlinear absorption is to reduce the intensity at the exit face of the sample. In this case, the transmitted intensity is no longer sensitive to the beam distortion and the z-scan data is a function of only the nonlinear absorption. Under this condition, the total transmittance is expressed as

$$T(z,S=1)=\sum_{m=0}^{\infty}\frac{\left[\frac{\beta I_o L_{eff}}{1+z^2/z_o^2}\right]^m}{(m+1)^{3/2}} \quad (5)$$

Once an open aperture z-scan measurement is performed, β can be determined.

3. Experiment

The study was conducted in two phases using Time-bandwidth's SESAM (Semiconductor Saturable Absorber Mirror) modelocked laser (wavelength, λ = 1064nm, average power, P_{avg} =1.64W, pulsewidth, τ_p =10ps) and repetition rate of 76MHz. The first phase comprised of measurements involving the use of the full 76MHz repetition-rate of the laser. The second phase involved measurements at a reduced laser repetition frequency using Quantum Technology Inc. Pockels cell pulse picker which enabled the repetition-rate of the laser to be reduced from 76MHz to 152Hz, with an opening rise time of ~ ns. The contrast ratio of extinguished to picked pulses was 250:1. A 2mm light-path pristine spectroscopic grade fused-quartz cuvette was used for both experimental arrangements and the light from the laser was focused using a 7.5cm focal length lens.

A LabVIEW-controlled stepper motor was used to move the sample within the focal plane. For the CA measurement, the transmitted intensity was measured as a function of the distance through which the sample was moved with respect to the focal plane. The beam was mechanically chopped and the output of the chopper was used as a reference signal for the lock-in amplifier. The detection scheme consisted of a silicon photodetector whose output was coupled into the lock-in amplifier.

4. Results and Discussion

Figure 2 shows a plot of CA z-scan measurement data for CS_2 at the full (high) repetition-rate of the laser and reduced (pulse-picked) repetition-rate.

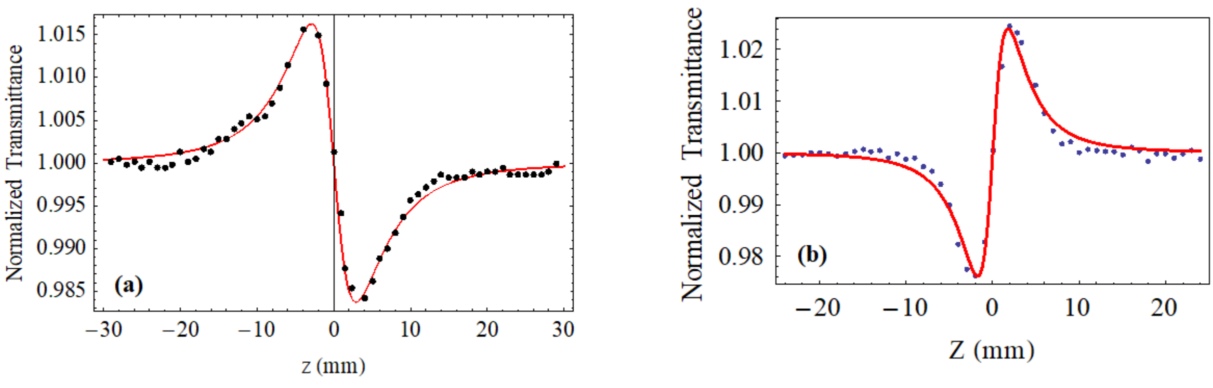

Figure 2. Closed aperture z-scan measurement of CS_2 using 10ps and 1.064μm laser pulses (a) at laser repetition-rate of 76MHz (b) Pockels cell pulse-picked repetition rate of 152kHz

Contrary to the expected valley-peak CA z-scan signature profile for CS_2 (a material with positive n_2), Figure 2(a) rather depicts peak-valley configuration and the n_2 was determined to be 1.1 x 10^{-12}cm/W which shows an order of magnitude enhancement at 76MHz. On the other hand, at a reduced repetition rate of 152MHz using Pockels cell pulse picker, the z-scan profile exhibited the expected configuration consistent with the behavior of CS_2 as depicted in Figure 2(b) and $n_2 = (2.9 \pm 0.2)\text{x}10^{-14}$W/cm.
In view of the measured high nonlinear index of refraction of CS_2 at 76MHz which could probably be due to thermal effects (Ganeev et al., 2004), the cuvettes were imaged using a Normaski microscope for any sign of possible damage. The results are depiced in Figure 3 in which (a)-(c) show the residual manufacturing defects

left on the surface of the glass through polishing (Genin, Feit, Kozlowski, Rubenchik, Salleo, & Yoshiyama, 2000; Honig, Norton, Hollingsworth, Donohue, & Johnson, 2005; Salleo, Genin, Yoshiyama, Stolz, & Kozlowski, 1998). On the other hand, Figure 3(d)-(f) show microexplosions triggered by irradiation of surface contaminants of the fused-quartz cuvettes using the high pulse repetition-rate laser (Genin, Feit, Kozlowski, Rubenchik, Salleo, & Yoshiyama, 2000; Honig, Norton, Hollingsworth, Donohue, & Johnson, 2005; Salleo, Genin, Yoshiyama, Stolz, & Kozlowski, 1998; Genin, Michlitsch, Furr, Kozlowski, & Krulevitch, 1997).

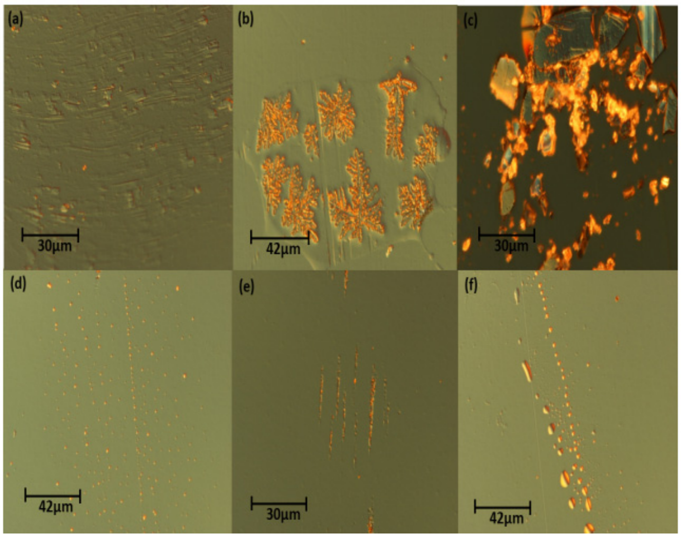

Figure 3. Normaski microscope images of damage morphologies of fused-quartz cuvette produced by 10ps, 1064nm and 76MHz laser pulses. (a)-(c) are residual defects left on the surface of the cuvettes during polishing (d)-(f) show microexplosions resulting from surface imperfections of the cuvettes

To further investigate the damage results shown in Figure 3, open aperture z-scan studies were carried out using empty damaged fused-quartz cuvettes at both 76MHz and the pulse-picked repetition rate of 152Hz and the results are displayed in Figure 4.

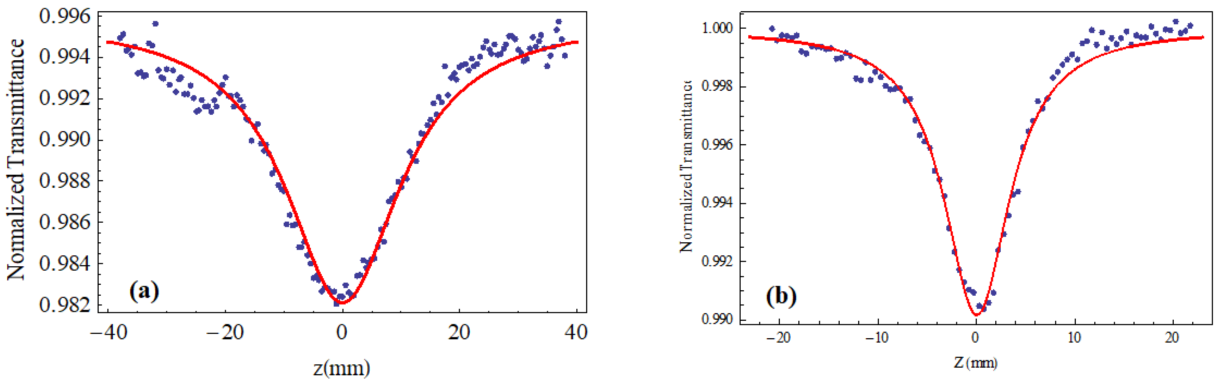

Figure 4. Open aperture z-scan results for a piece of empty LID fused-quartz at (a) 76MHz resulting in $\beta = (6.9 \pm 0.2)\times10^{-8}$cmW^{-1}, which shows two orders of magnitude enhancement and (b) pulse-picked frequency of 152Hz for which $\beta = (6.9 \pm 0.2)\times10^{-8}$cmW^{-1}

Based on Figure 4(a), the nonlinear absorption coefficient for LID fused quartz at 76MHz and at 1064nm is $(6.9 \pm 0.2)\times10^{-8}$cmW^{-1}, which shows two orders of magnitude enhancement over the literature value of $(7.5 \pm 0.4)\times10^{-10}$cmW^{-1} within the picosecond regime (Repeev, 1994). On the other hand, Figure 4(b) depicts open aperture results at 152 Hz resulting in $\beta=(1.3 \pm 0.2)\times10^{-10}$cmW^{-1}. Thus, the strong two-photon absorption observed at 76MHz was triggered by microexplosions resulting from plasma formation at the manufacturing-induced defect sites. Interference between light from these damaged sites and the actual z-scan signal modifies the profiles resulting in the enhanced measured nonlinear absorption coefficient. Thus, such orders of magnitude enhancement of the nonlinear absorption coefficient of fused-quartz in high repetition-rate z-scan coupled with images of the damage morphologies of these materials could be a timely indicator of LID.

5. Conclusion

The study examined the effect of high repetition-rate laser on the nonlinear response of a standard calibration material, CS_2 and contained in fused-quartz cuvettes. The results show that as compared to low repetition-rates results, high laser repetition rate closed aperture z-scan profiles are distorted and the measured nonlinear index of refraction of is enhanced due to thermal effects. The study also established that at high laser repetition rates and depending on the nature of the surfaces involved, apparently high LID materials do experience below-threshold damage and the response of these damaged materials do affect the overall transmitted z-scan signal; Normaski microscopy images confirmed the damage. Based on these results, a criterion for timely recognition of LID in ultrashort high-repetition-rate z-scan measurements has been formulated to help avoid erroneous interpretation of the origin and strength of nonlinear response in such studies. For the first time, a z-scan study has successfully tied LID to laser repetition-rate.

Conflict of interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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