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Simultaneous measurement of pure-optical and thermo-optical nonlinearities induced by high-repetition-rate, femtosecond laser pulses: application to CS_2

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Abstract. A simple experimental technique is presented capable of separating the contribution of purely optical Kerr effects from that of thermo-optical effects in the nonlinear response of materials under high-repetition-rate laser irradiation. The technique has been realized by combining the single-beam *Z*-scan method with the single-beam thermal lens measurement method. We demonstrate this technique by analysing the nonlinear response at 770 nm of CS_2 which exhibits cumulative thermal effects when irradiated by very intense femtosecond laser pulses at a 76-MHz repetition rate.

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All-optical switches operating at high pulse-repetition rates are the basis for developing optical parallel data processing systems and optical time domain multiplexing transmission systems. High switching speeds, however, have to be coupled with low switching energy and ultrafast recovery times, and these requirements could be met by materials with enhanced nonresonant optical nonlinearities. Nevertheless operation at very high pulse-repetition frequencies may give rise to cumulative effects capable of completely modifying the nonlinear behaviour of even these nonresonant materials. In fact, at these operating regimes, the properties of the transmitted light are determined both by the linear and nonlinear optical response of the materials, and by possible thermal effects caused by even very low single or multiple photon absorption of the laser energy. In evaluating the potentialities of a nonlinear material for applications requiring operation at high repetition rates one has to use experimental methods capable of separating the contribution of the purely optical Kerr effect from that of the thermo-optical effects. Among these methods, a relatively simple one can be realized by combining the single-beam *Z*-scan technique for the determination of sign and magnitude of third-order optical nonlinearities [1], with the single-beam thermal lens measurement technique, first developed for thermal lens spectroscopy [2]. Here we describe how this can be done and we demonstrate the "combined" technique potentialities by applying it to CS_2, a solvent that is frequently used as a standard reference nonlinear material [1]. In fact we have noticed that under relatively intense high-repetition-rate fs laser irradiation, the overall CS_2 nonlinear response may be substantially affected by slow thermal nonlinearities due to cumulative effects.

The schematic diagram of the experimental arrangement is shown in Fig. 1. Laser source is a self-mode-locked Ti:sapphire laser delivering 130-fs-long, linearly polarized pulses with 76-MHz repetition rate and peak powers up to 125 kW. Measurements are performed at a laser wavelength of 770 nm. The Ti:sapphire beam is focused by a set of three converging lenses (focal lengths: 10, 9, and 7.5 cm). In our experiment, the laser beam waist spot size at the focal plane is 25 ± 1 μm. Exposure of the sample to the excitation beam is controlled by a chopping wheel, which is put at the focal plane of lens 1 to minimize the spot size of the excitation laser on the wheel. Chopper frequency is set at 14 Hz for a chop-

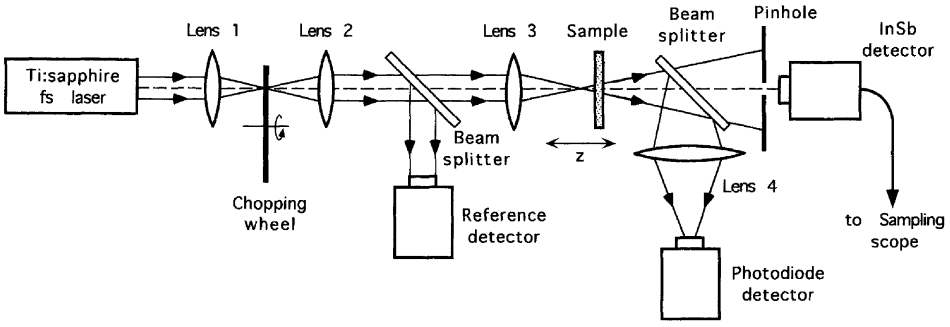

Fig. 1. Experimental arrangement for combined single-beam *Z*-scan and thermal lens measurements

ping wheel duty cycle that depends on the width of the wheel window. The chopper opening risetime is approximately 15 μs. The experimental set-up allows simultaneous monitoring of the near-field beam intensity transmitted through the sample, and of the far-field light transmission at the sample exit through a 0.5-mm-diameter pinhole placed at a distance of more than 60 times the diffraction length away from the focal plane. The transient behaviour of the transmitted far-field light is detected by a fast InSb detector whose output is integrated, displayed and recorded by a digital averaging oscilloscope (Tektronix TDS 520A) set at 20 MHz bandwidth and at twenty times average mode. This permits us to monitor the time evolution of the envelope of trains of pulses whose overall temporal duration is determined by the chopper opening interval. Measurements are carried out by recording magnitude and temporal profile of the far-field-transmitted on-axis signal as a function of the position of the sample around the focal plane. In this way the measured quantity is the sample transmittance as a function of time during the time interval when the excitation beam is unobstructed (thermal lens measurement), and as a function of the sample position measured with respect to the focal plane (Z-scan). It is therefore possible to follow the time evolution of the Z-scan traces from 15 μs (the chopper opening risetime) onwards.

In performing third-order susceptibility measurements with the Z-scan technique, it is customary to use CS_2 as a reference material. For this reason, we have tested the "combined" technique on this material. Liquid CS_2 was enclosed in a 1-mm fused quartz cell with wall thickness of 1.25 mm, placed on a microcomputer-controlled mechanical translator. Using this cell, the sample thickness was shorter than the focused beam diffraction length in the interaction region. In our experiment we have chosen a duty cycle of 0.07, which implies that the sample is exposed to the excitation laser for 5 ms at intervals of about 71 ms. Under these experimental conditions, Z-scan traces with a peak-to-valley separation larger than the technique resolution (i.e. $> 1\%$) were observed only for peak irradiances exceeding 1.5 GW/cm^2. At these levels it was evident that the transmitted beam was affected by thermal lensing. This is demonstrated by the time evolution of the transmitted signal, indicative of the build-up of a thermal lens effect, as shown by the typical set of recorded oscilloscope traces reported in Fig. 2. Here, we report traces recorded at a peak irradiance of 12.5 GW/cm^2 in order to visually emphasize the effect. Analysis of these traces reveals that the time constants of the thermal effect are consistent with the theoretical values of the time required for thermal conduction effects to become significant, t_c, defined as $t_c = \omega_0^2 \rho c_p / 4\kappa$ [3], where ω_0 is the laser beam radius at the sample, $\rho = 1.263\,\mathrm{g\,cm^{-3}}$ the CS_2 density, $c_p = 1.045\,\mathrm{J\,g^{-1}K^{-1}}$ its specific heat, and $\kappa = 160\,\mathrm{mW\,m^{-1}K^{-1}}$ its thermal conductivity [4, 5]. In fact, under the irradiation conditions characterizing our experiment, building of the thermal lens is dominated by thermal conduction effects rather than by acoustic density changes [6]. In our case, the theoretical value of t_c varies between 1.3 ms and 2.6 ms for laser beam radii comprised within a diffraction length from the focal plane.

Typical examples of the Z-scan curves obtainable at different times from each set of oscilloscope traces are shown in Fig. 3. It is evident that at the earliest times after illumination of the sample begins, the Z-scan trace exhibits the peak–valley profile characteristic of a positive nonlinear refractive index, as expected for CS_2 at this wavelength [1]. Here, thermal effects are not capable of overwhelming the nonlinear effects of electronic origin owing to the relatively slow thermal lens build-up time. In contrast, at later times, i.e. 150–200 μs later, the peak–valley profile turns upside down, with a behaviour characteristic of thermal self-defocusing in a medium with a negative temperature coefficient of the refractive index, dn/dT, as is the case for CS_2. Thermal nonlinearities have

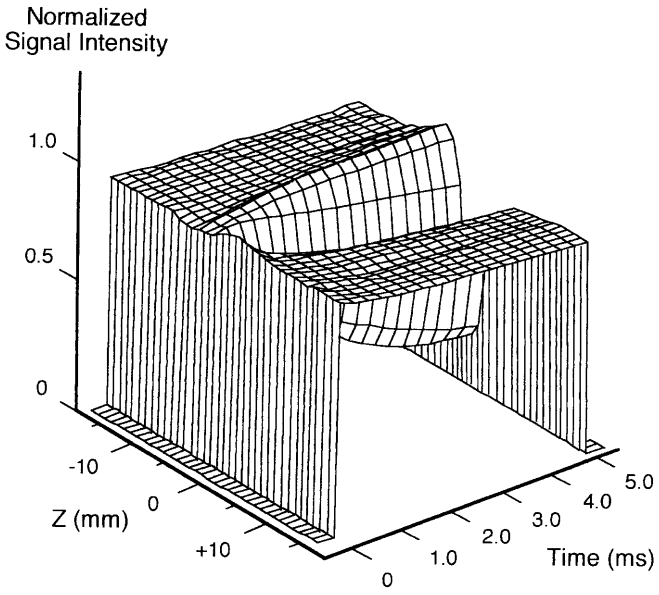

Fig. 2. Set of oscilloscope traces showing the normalized signal intensity as a function of CS_2 position with respect to the focal plane. Peak irradiance equal to 12.5 GW/cm^2

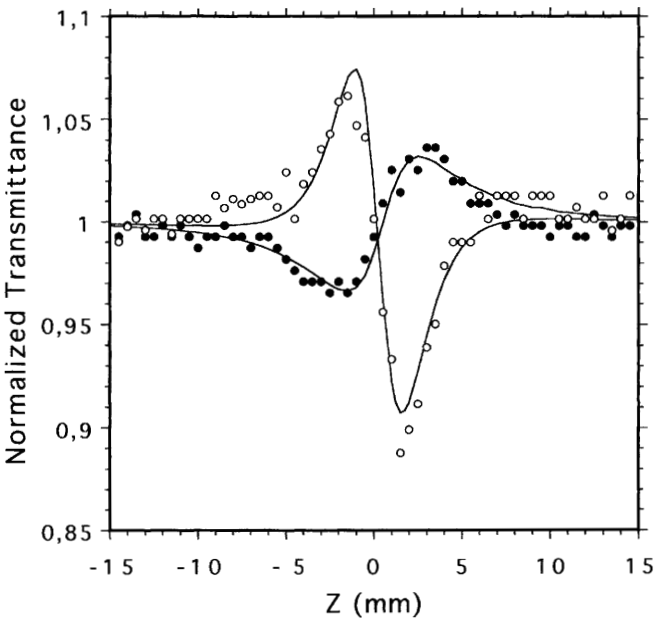

Fig. 3. Normalized transmittances of the Z-scan measurements from a 1-mm-thick CS_2 cell at a peak irradiance of 12.5 GW/cm^2. *Filled points*: Z-scan immediately at the end of the chopper opening risetime. *Open points*: Z-scan 400 μs later. The *solid lines* are theoretical calculated results, as discussed in the text

already been observed in CS_2 for irradiation with TEA CO_2 laser single pulses having lengths from 30 to 300 ns [1, 7]. These nonlinearities are caused by the sizeable linear absorption of CS_2 at 10.6 μm, with a coefficient $\alpha \approx 0.22\,\mathrm{cm}^{-1}$ [1]. However, linear absorption at 770 nm is expected to be much lower (a value of $\alpha \approx 1.4 \times 10^{-4}\,\mathrm{cm}^{-1}$ has been measured at 632 nm [4]) and, therefore, is most likely insufficient to account for the magnitude of the observed thermal effects. Indeed, we have made an attempt to measure the possible thermal lens contributions due to linear absorption by operating the laser source in a cw mode. At the highest emitted power, we have observed a residual thermal lens effect with a maximum difference between the normalized peak and valley transmittance, $\Delta T_{\mathrm{p-v}}$, in the Z-scan trace at 5 ms equal to approximately 10%. This value is almost one order of magnitude lower than the corresponding value measured when the laser is mode-locked. The small thermal lens effect observable under cw conditions could be due either to the cell walls or to CS_2.

An additional evidence that thermal effects cannot be attributed to linear absorption has been derived by following the $\Delta T_{\mathrm{p-v}}$, time evolution for Z-scan curves recorded at different irradiances of the mode-locked excitation beam. Typical results are reported in Fig. 4, for two values of peak irradiance, i.e. 7.5 and 12.5 GW/cm^2. The value of $\Delta T_{\mathrm{p-v}}$, for Z-scans immediately at the end of the chopper opening risetime is seen to increase linearly with peak irradiance, as one should expect for a cubic (n_2-type) nonlinearity. At later times, where thermal effects dominate as shown by the reversed valley–peak pattern of the Z-scan curves, $\Delta T_{\mathrm{p-v}}$ is seen to vary quadratically with peak irradiance. Moreover, here the curve peak and valley become closer, with a separation approaching $1.2z_{\mathrm{O}}$, where z_{O} is the focused beam diffraction length, as compared to $1.7z_{\mathrm{O}}$ obtained for the cubic nonlinearity. It has to be noted that results previously reported in literature for thermal effects connected with linear absorption normally show a separation larger than the $1.2z_{\mathrm{O}}$ value obtained here [8, 9]. Both our later-times evidences, i.e. the quadratic dependence of $\Delta T_{\mathrm{p-v}}$, and the reduced peak-to-valley separation, point to an effective higher-order effect resulting from a nonlinear absorption giving rise to the thermo-optical nonlinearity. A possible mechanism, even though not the only one, could be two-photon absorption. Indeed CS_2 has a reasonably intense absorption band in the 290–410 nm region [10] and multi-photon absorption in CS_2 is a process that has been widely exploited to study the excited states of this molecule [11]. Note that the thermal lens technique has been already applied in the past for the measurement of multiphoton absorption [12].

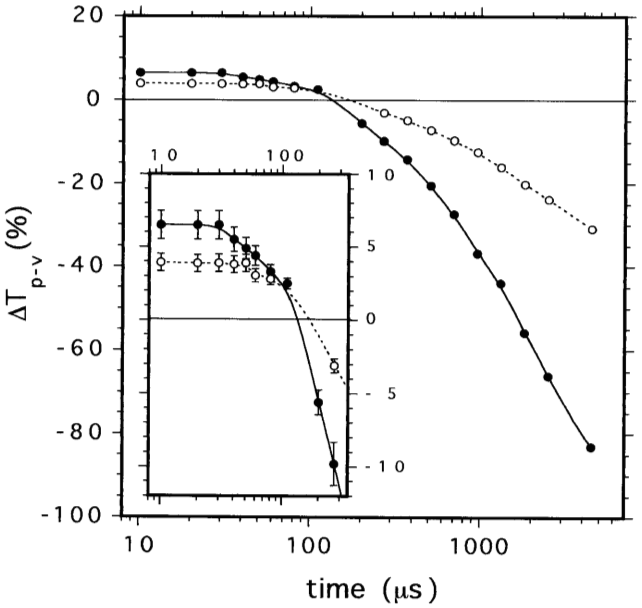

Fig. 4. $\Delta T_{\mathrm{p-v}}$ in percent as a function of time from the Z-scan data of CS_2 at a peak irradiance of 7.5 GW/cm^2 (*open points*) and 12.5 GW/cm^2 (*filled points*). The *inset* shows the behaviour of $\Delta T_{\mathrm{p-v}}$ during the initial 300 μs with an expanded vertical scale

We have also tried to observe directly the effects of nonlinear absorption by measuring the near-field unapertured transmittance with the beam-splitter, lens 4, and photodiode detector assembly shown in Fig. 1. However no transmittance change with position was detected for excitation up to the highest irradiances, indicating that if nonlinear absorption processes exist, their effects on the open-aperture transmittance are beyond the sensitivity of our open-aperture Z-scan apparatus. Owing to the impossibility of directly measuring the value of the nonlinear absorption coefficient β, its value has been obtained by least-square fits to each Z-scan curve using a theoretical model based on those usually employed to analyze the Z-scan data [1]. In our case the temperature field produced in the sample is described by a two-dimensional temperature model [3] considering both linear and nonlinear absorption, and Huygens's integral is used to evaluate the propagation of the laser field from the sample to the detector plane. By using this model we have obtained a value of the nonlinear absorption coefficient $\beta = (4.5 \pm 1.0) \times 10^{-4}$ cm/GW, and a value of the nonlinear refractive index $\gamma = 2.3 \times 10^{-6}\,\mathrm{cm}^2/\mathrm{GW}$ ($n_2 = 8.9 \times 10^{-13}$ esu), this last value with an uncertainty of a factor of two. This factor arises essentially from the relatively high noise of the data recorded at the earliest times, from the uncertainty in the peak irradiance values (i.e. beam waist, pulsewidth, and energy calibration), and from that associated with the fitting procedure. The relatively low value of β makes it evident that a large nonlinear absorption is not required for cumulative thermal effects to become significant. This demonstrates why care has to be taken in performing conventional Z-scans with high-repetition-rate lasers: in this case the results of measurements carried out even at very low average powers by averaging over long time intervals might be incorrectly interpreted for the presence of cumulative phenomena due to absorption processes that the standard measurement techniques are not able to uncover. At the highest peak irradiances of our experiment, thermal effects would become negligible only for repetition rates lower than a few hundred kHz, as we have verified by simulating the CS_2 response by means of the computer model used for deriving the CS_2 nonlinear coefficients.

As far as the CS_2 nonlinear refractive index n_2 is concerned, it has to be noted that it has already been measured under fs excitation using (i) time-resolved interferometry at 630 nm with a 100-fs laser system at 10 Hz, yielding a value of 3×10^{-12} esu with an uncertainty of a factor of two [13], and (ii) beam-deflection methods at 616 nm with a 75-fs, low repetition-rate laser system yielding a value of $(1.9 \pm 0.6) \times 10^{-12}$ esu [14]. These values are from four to ten times smaller than those measured using ps or longer laser

pulses, ranging from 11×10^{-12} to 20×10^{-12} esu [15]. This is due to the fact that only the instantaneous electronic and the fast molecular libration processes contribute to the nonlinear response under fs irradiation since molecular reorientation and redistribution are significantly slower processes [16]. The value of n_2 obtained from our measurements agrees with those reported in [13] and [14] once the uncertainties are taken into account.

In conclusion, the results of our measurements on CS_2 demonstrate that the combined technique utilizing a high repetition-rate laser makes it possible to retrieve at the same time the sample nonlinear refractive index for fs laser irradiation, and its nonlinear absorption coefficient. Measurements performed at very high repetition rates may be relatively simpler and faster than those performed in single shot, as is customary in the conventional closed-aperture Z-scan technique for reducing possible thermal effects. An additional advantage of the combined technique resides in its enhanced sensitivity as compared to the conventional open-aperture Z-scan technique. This makes the technique suitable if one wants either to measure very low nonlinear absorption coefficients, otherwise unmeasurable, or to check for the presence and relevance of thermo-optical effects.

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