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Z-scan technique through beam radius measurements

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ABSTRACT A modification to the well-known *z*-scan technique for measuring optical non-linearities is introduced. It is based on directly measuring the beam radius in the far field instead of the transmittance of the irradiance through an aperture, as in the original version. It has the advantage of being insensitive to beam pointing instability and is almost insensitive to power fluctuations. Furthermore, the calculations required for the determination of the non-linear parameters are simplified. For demonstrating the advantages of the modified method, beam radius and transmittance measurements were simultaneously taken in the standard non-linear optical material CS_2. Separate fittings of these measurements gave almost the same values for the non-linearities, quite similar to those in the literature. A common fitting has been applied to both sets of measurements, enhancing the accuracy of the method.

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1 Introduction

In 1989, Sheik-Bahae et al. [1] reported a method, the *z*-scan technique, for the determination of the sign and magnitude of the third-order non-linear refractive index γ in optical materials. One year later, the same authors described this method extensively and demonstrated how it can be applied to a variety of materials [2]. Ever since, the *z*-scan has been the most widely used technique for measuring the non-linear optical properties of materials because of its experimental simplicity and ease of use.

In this technique [2], a sample is scanned along the optical axis (*z*-direction) of a focused Gaussian laser beam, around its focus. The high intensity of the electromagnetic field in this region induces on the sample a non-linear lens of variable focal length. This causes a refractive divergence or convergence of the laser beam, depending on the sample position relative to the beam focus and the sign of the non-linearity. The resulting variations in the beam radius in the far field are able to give adequate information for the calculation of the refractive non-linearities. Therefore, the beam radius is the most fundamental parameter for the determination of the non-linear refractive index of the material. Nevertheless, in the original version of the *z*-scan technique, the measured quantity is not the beam radius but a derivative parameter of it, the normalized transmittance of the irradiance through a finite hard aperture in the far field [2]. However, the measurement of this parameter entails disadvantages, such as high sensitivity in beam pointing instability and power fluctuations [3, 4], as well as complexity in the numerical calculations. Several modifications of the *z*-scan technique have been proposed to overcome its deficiencies and increase its capabilities. Top-hat beam [5, 6] and eclipsing *z*-scan [7] have been used to enhance the sensitivity of the technique. Also, thick optical materials [8–10] have been utilized to increase the measured signal. In a further development, the use of a CCD camera increased the accuracy of the method [4, 11]. It is worth emphasizing that in all these modifications, the measured parameter is the normalized transmittance through a hard or soft aperture.

In the present work, a fundamental modification of the *z*-scan technique concerning the measured parameter is introduced. We propose the direct measurement of the beam radius in the far field through a CCD camera connected to a laser beam profiler. This modification has the advantage of being insensitive to pointing instability, because no hard aperture is employed, and is almost insensitive to power fluctuations, because the beam radius does not depend on the total power of the beam but only on its spatial distribution. In practice, the latter eliminates the need for using a reference beam, as occurs in the case of the transmittance *z*-scan, simplifying the experimental set-up. In addition, the numerical calculations involved in this modification are simplified since we directly measure the beam radius and not a derivative parameter of it, such as the transmittance through an aperture. Besides, the combination of the CCD camera with the beam profiler provides a continuous supervision of the entire beam profile during the experiment.

2 Theoretical aspects

The electric field pattern of a circular Gaussian beam, passed through a thin sample of a non-linear material, can be obtained by the Gaussian decomposition method [2] and, at a distance *d* from the sample, is given by the relation-

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$$E_{\mathrm{d}} = (z, r, t;\, \Delta\Phi_0(t)) = E(z, r=0, t)\mathrm{e}^{-\alpha L/2} \times \sum_{m=0}^{\infty} \frac{[i\Delta\Phi_0(z,t)]^m}{m!} \frac{w_{m0}}{w_m} \exp\left(-\frac{r^2}{w_m^2} - \frac{ikr^2}{2R_m} + i\theta_m\right). \quad (1)$$

Here, z is the distance between the sample and the beam waist, defined negative if the sample is in the front of the waist (Fig. 1a), $E(z, r, t)$ is the electric field pattern of the incident Gaussian beam at the sample plane, k is the wave number, α the linear absorption coefficient and L is the sample thickness. If we define

$$d_m = \frac{k w_{m0}^2}{2}, \quad (2)$$

and

$$g = 1 + \frac{d}{R(z)}, \quad (3)$$

the remaining parameters are given by the relationships

$$w_{m0}^2 = \frac{w^2(z)}{2m+1}, \quad (4)$$

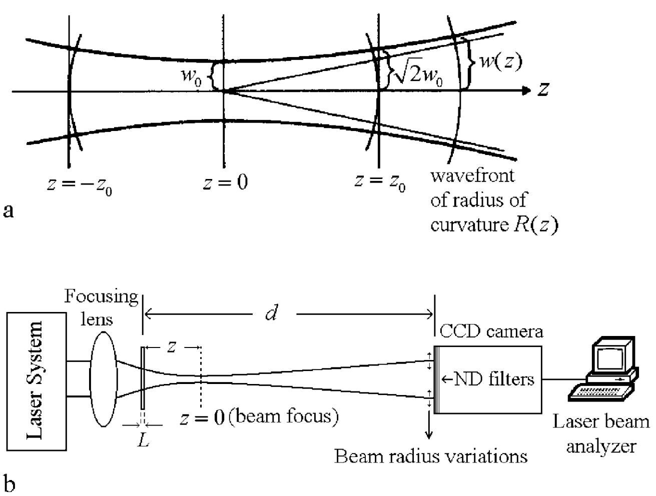
$$w_m^2 = w_{m0}^2\left[g^2 + \frac{d^2}{d_m^2}\right], \quad (5)$$

$$R_m = d\left[1 - \frac{g}{g^2 + d^2/d_m^2}\right]^{-1}, \quad (6)$$

and

$$\theta_m = \tan^{-1}\left[\frac{d/d_m}{g}\right], \quad (7)$$

where $w(z)$ is the beam radius and $R(z)$ the radius of the wave front curvature of the incident beam at the sample plane (Fig. 1a), while d is the distance between the sample and the detection plane (Fig. 1b).



b

FIGURE 1 **a** The fundamental parameters of a Gaussian beam. **b** Experimental set-up for the application of the proposed technique

The parameter $\Delta\Phi_0(z, t)$ is the on-axis phase shift induced to the beam by the refractive non-linearities of the material, and it is related to the refractive index change at the focus $\Delta n_0(t)$ by the relationship

$$\Delta\Phi_0(z, t) = \frac{k\Delta n_0(t) L_{\mathrm{eff}}}{1 + z^2/z_0^2}, \quad (8)$$

where z_0 is the Rayleigh length of the beam and L_{eff} the effective propagation length inside the sample, defined as $L_{\mathrm{eff}} = [1 - \exp(-\alpha L)]/\alpha$. $\Delta\Phi_0(t)$ is the on-axis phase shift at the focus ($z = 0$). Finally, the refractive index change $\Delta n_0(t)$ is related to the non-linear refractive index γ by the relationship

$$\Delta n_0(t) = \gamma I_0(t), \quad (9)$$

where $I_0(t)$ is the on-axis value of the irradiance at the focal point.

In the original version of the z-scan technique, the measured quantity was the normalized transmittance, defined by the relationship [2]

$$T(z) = \frac{\int_{-\infty}^{+\infty} P_{\mathrm{T}}(z, \Delta\Phi_0(t))\mathrm{d}t}{S\int_{-\infty}^{+\infty} P_{\mathrm{i}}(t)\mathrm{d}t}, \quad (10)$$

where $P_{\mathrm{i}}(t)$ is the instantaneous input power (within the sample), S the aperture linear transmittance and $P_{\mathrm{T}}(z, \Delta\Phi_0(t))$ the transmitted power through the aperture obtained by spatially integrating the irradiance up to the aperture radius r_{a}, i.e.

$$P_{\mathrm{T}}(z, \Delta\Phi_0(t)) = c\varepsilon_0\pi \int_0^{r_{\mathrm{a}}} |E_{\mathrm{d}}(z, r, t;\, \Delta\Phi_0(t))|^2 r\mathrm{d}r. \quad (11)$$

In the above equation, c is the speed of light in a vacuum, and ε_0 is the vacuum permittivity.

By fitting (10) to the experimental z-scan measurements of the transmittance, the phase parameter $\Delta\Phi_0(t)$ and consequently the refractive index change $\Delta n_0(t)$ as well as the non-linear refractive index γ can be calculated.

In contrast, in the proposed modification the quantity measured is the beam radius r_q. This is defined as the distance from the beam center to the points where the irradiance reduces to a specific fraction q of its on-axis ($r = 0$) value. For example, a value of q equal to $1/\mathrm{e}^2 = 0.1353$ corresponds to the commonly defined radius $w(z)$ of the Gaussian beam.

The radius is numerically calculated through the relationship

$$I(z, r_q, t;\, \Delta\Phi_0(t)) = \frac{c\varepsilon_0}{2}|E(z, r_q, t;\, \Delta\Phi_0(t))|^2 = qI(z, 0, t;\, \Delta\Phi_0(t)). \quad (12)$$

The value of the non-linear phase shift $\Delta\Phi_0(t)$ and consequently the refractive index change $\Delta n_0(t)$ as well as the non-linear refractive index γ is calculated by fitting (12) to the experimental z-scan measurements of the radius. Obviously, the numerical calculations involved in the proposed modification are much simpler than those required in the original version of the z-scan technique, mainly because the spatial integration of (11) is avoided.

3 Experiment, calculations and discussion

The experimental set-up of the proposed technique is shown in Fig. 1b. The variations in the beam radius were measured through a laser beam profiler (Spiricon, Inc. LBA-300 PC Laser Beam Analyzer). The detection system of this instrument is based on a two-dimensional CCD camera (Cohu, Inc. 4915 RS-170 CCD camera) having 768×494 pixels (pixel size $8.4\,\mu\text{m} \times 9.8\,\mu\text{m}$). The profile of the beam, together with the beam parameters, were automatically displayed on a computer screen, giving the values of the beam parameters simultaneously. The fluctuations in the measurements were adequately suppressed by averaging the values of 100 measurements collected by the instrument. The standard non-linear material CS_2 contained in a cell of $L = 1$ mm thickness was used as a sample. A mode-locked Ti: sapphire femtosecond system (TSUNAMI – Spectra Physics) pumped by a continuous-wave (CW) frequency-doubled Nd:YVO_4 laser (MILLENNIA – Spectra Physics) emitting approximately 85-fs pulses at 800 nm was utilized as the excitation source. The laser beam was focused on a $w_0 \approx 55$-µm-radius waist, producing a peak value of the on-axis irradiance at focus $I_0 = 2.4\,\text{GW/cm}^2$. The beam profile was found to be very close to a circular Gaussian one. The Gaussian profile is attributed to the fact that the mode-locking process in the laser by itself optimizes the beam to be an almost ideal Gaussian TEM_{00} mode [12]. Beam radius measurements taken simultaneously with transmittance measurements are shown in Figs. 2 and 3, respectively. For the beam radius measurements, a value of q equal to 0.08 was chosen as the optimum for our specific experimental conditions, while, for the transmittance measurements, a soft aperture with linear transmittance $S = 0.4$ was used. The radius and transmittance measurements were normalized with respect to their values in the linear regime (far from the focus).

Because of the high repetition rate of the laser system (82 MHz), the thermal non-linearities of the material are dominant. Due to the much longer relaxation time of the induced thermal lens in CS_2 (~ 100 ms) [13] as compared with the temporal separation of the pulses (~ 12.2 ns), the sample was considered to be in a quasi-steady state. Under these conditions, the average values of the non-linear optical parameters were calculated. The separate least-square fittings of the beam radius and transmittance measurements (Figs. 2 and 3) gave $\langle\Delta\Phi_0\rangle = -0.78$ and -0.68, respectively. These values implied a refractive index change at the focus $\langle\Delta n_0\rangle = -9.9 \times 10^{-5}$ for the beam radius fitting and $\langle\Delta n_0\rangle = -8.7 \times 10^{-5}$ for the transmittance fitting. In order to compare our results with those reported in the literature, an equivalent refractive index $\gamma_{\text{eq}} = \langle\Delta n_0\rangle / I_0$ for the thermal non-linearities has been used. In our case, the values of γ_{eq} were $-4.1 \times 10^{-14}\,\text{cm}^2/\text{W}$ and $-3.6 \times 10^{-14}\,\text{cm}^2/\text{W}$ for the radius and transmittance measurements, respectively. The reasonable agreement of the γ_{eq} values for the two cases verifies the validity of the suggested modification to the *z*-scan technique. Also, these values are quite similar to those reported in the literature [14], within the experimental errors originating mainly from the uncertainty in the determination of the peak value of the on-axis irradiance at the focus I_0 (i.e. the determination of the beam waist, pulse width and energy calibration).

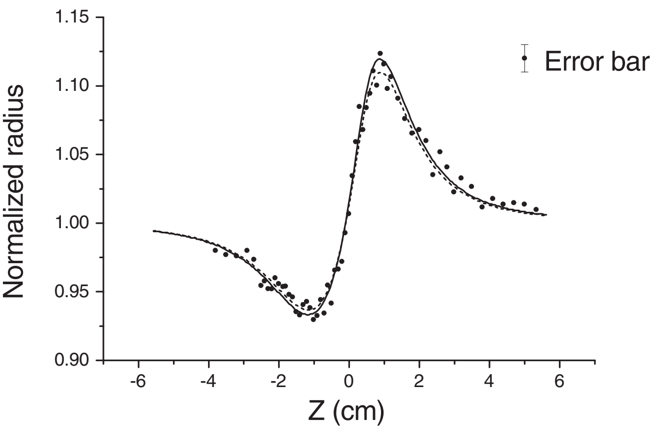


FIGURE 2 Radius *z*-scan plot for a 1-mm-thick CS_2 cell. The valley–peak configuration is characteristic of self-defocusing induced by thermal effects. The *solid* and *dashed lines* correspond to the separate ($\langle\Delta\Phi_0\rangle = -0.78$) and common ($\langle\Delta\Phi_0\rangle = -0.73$) fittings, respectively. The value of the parameter q chosen was 0.08. Both fittings were performed with $w_0 = 53.5\,\mu\text{m}$ ($z_0 = 1.124$ cm)

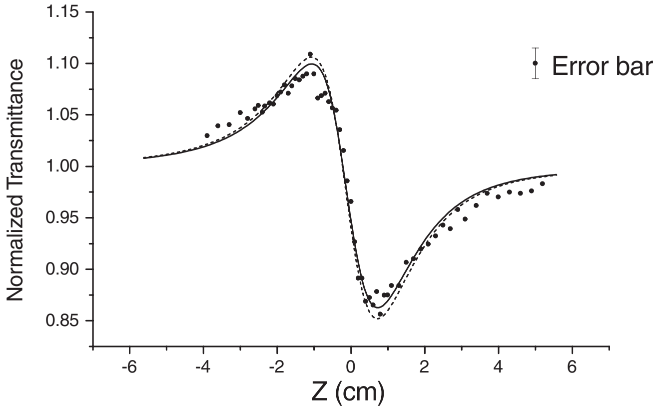


FIGURE 3 Transmittance *z*-scan plot simultaneously taken with the radius plot. The peak–valley configuration is again characteristic of thermal self-defocusing. The *solid* and *dashed lines* correspond to the separate ($\langle\Delta\Phi_0\rangle = -0.68$) and common ($\langle\Delta\Phi_0\rangle = -0.73$) fittings, respectively. The value of w_0 used was again $53.5\,\mu\text{m}$ ($z_0 = 1.124$ cm)

Numerical simulations based on (12) prove that for $|\Delta\Phi_0| \leq 1$, the difference between the normalized peak and valley radii $\Delta r_{\text{P-V}}$ in a radius *z*-scan plot depends almost linearly on $|\Delta\Phi_0|$. Furthermore, a simplified relation connecting $\Delta r_{\text{P-V}}$ and $|\Delta\Phi_0|$ within an accuracy of $\pm 2\%$ for different values of q has been deduced. This is

$$\Delta r_{\text{P-V}} = 0.154 q^{-0.214} |\Delta\Phi_0| \,. \tag{13}$$

This relationship can be used to readily estimate the non-linear refractive index γ by simply measuring the normalized peak–valley difference. Also, (13) provides a measure of the sensitivity of this technique, defined as [1, 7]

$$p = \Delta r_{\text{P-V}} / |\Delta\Phi_0| = 0.154 q^{-0.214} . \tag{14}$$

Its dependence on the parameter q is shown in Fig. 4. As the value of q decreases, the sensitivity of the method increases dramatically, and it finally becomes considerably higher than that of the original *z*-scan version. This occurs because for

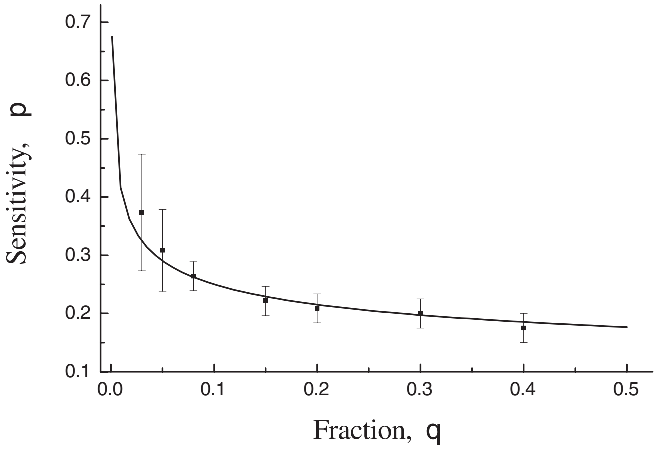


FIGURE 4 Sensitivity of the proposed z-scan technique versus the fraction q of the peak irradiance for which the beam radius is defined. The *square points*, together with the *error bars* depict the experimental measurements and their uncertainty

low values of q the beam radius lies in the region of the beam wings, and its variations become very large. However, for very low values of q, the beam intensity is comparable to the noise level, introducing high uncertainty into the measurements. Therefore, the value of q must be kept at an optimum level in order to achieve a reliable signal-to-noise ratio. In our case, this value was 0.08, as mentioned above. Z-scan measurements of the radius were also realized for different q values, giving similar results concerning the non-linear parameters. The sensitivity of each case was estimated through (14) and its values are depicted in Fig. 4 (square points), verifying the validity of (13). As can be seen in this figure, for values of q lower than 0.08, the noise strongly influences the measurements, indicated by the long error bars. For $q > 0.08$, the uncertainty in the measurements is suppressed. Therefore, at the optimum value of $q = 0.08$ we achieve a compensation between high sensitivity and low noise.

Another important advantage of using a beam profiler is the simultaneous measurement of different parameters of the beam, such as the radius and the transmittance through a soft aperture, as it was reported previously. The simultaneity of the measurements secures identical experimental conditions, permitting the performance of a common fitting, i.e. a fitting of the radius and transmittance measurements with common values of the parameters w_0 and $\langle \Delta\Phi_0 \rangle$. Especially, the transmittance measurements are fitted through (10) and (11), while the radius measurements are fitted through (12), both employing the same electric field distribution $E_\mathrm{d}(z, r, t; \langle \Delta\Phi_0 \rangle)$ (1). The common parameters w_0, $\langle \Delta\Phi_0 \rangle$ are chosen so that both sets of measurements are best fitted. The common fitting improves the accuracy of the method, since the data originating from two different experimental procedures are simultaneously taken and fitted. In our case, this common fitting is shown by the dashed lines in Figs. 2 and 3, giving $\langle \Delta\Phi_0 \rangle = -0.73$, which entails $\langle \Delta n \rangle = -9.3 \times 10^{-5}$, and finally $\gamma_\mathrm{eq} = -3.85 \times 10^{-14}\ \mathrm{cm^2/W}$. Strictly speaking, the results of the common fitting are equivalent to the average of the results obtained by the two separate fittings, taking into account the accuracy of each measuring procedure as a statistical weight. Especially, in our case the accuracy of the two measuring procedures, as can also be deduced from the dispersion of the experimental points (Figs. 2, 3), is almost the same. Therefore the $\langle \Delta\Phi_0 \rangle$ value obtained by the common fitting coincides with the simple arithmetic mean of the values obtained by the separate fittings.

Finally, an extension of this method can be achieved by simultaneously performing close (radius–transmittance) and open-aperture z-scans with the same beam profiler. In this way, both the real and the imaginary part of the optical susceptibility can be measured by performing a single experiment and using only one CCD camera and beam profiler.

4 Conclusions

In conclusion, a modification to the z-scan technique, based on the direct measurement of the variations of the beam radius in the far field using a CCD camera and a laser beam profiler, has been proposed. As a demonstration, the method has been applied to measuring the thermal nonlinearities of CS_2. The validity of the proposed modification has been verified by simultaneously performing a transmittance z-scan using a soft aperture. The main advantages of the proposed method are the continuous supervision of the entire beam profile, the elimination of the problem of pointing instability and the suppression of the influence of the laser power fluctuations on the experimental data. Furthermore, the use of the beam profiler gives the opportunity of simultaneously performing z-scan measurements for different parameters (e.g. beam radius and transmittance through a soft aperture), allowing a more accurate estimation of the non-linear refractive index. Simultaneous performance of closed- and open-aperture z-scan experiments is also possible. Finally, the method can be easily extended to the very common case of a non-perfect circular Gaussian beam. In this case, the original technique of measuring the transmittance through a finite circular aperture is practically extremely difficult. This extension will be the subject of a forthcoming work.

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