the global average. This is reflected in the *X*-scores and *F*-scores (Tables 4 and 5). The NUS + NTU cluster is four times more effective than the IISc + 7IITs cluster at the level of the second-order indicators[10,11].

Finally, we can give a broad estimate for the balance (evenness or consistency) as measured by $\eta = F/X$ in the two clusters arranged into the boarder REF categories, as shown in Table 6. We see that the NUS + NTU cluster has a slight edge over the IISc + 7IITs cluster.

In conclusion, we decompose the research performance of the IISc + 7IITs and NUS + NTU clusters into three components – size, excellence and balance or evenness. Data are retrieved from the excellence mapping web application. The NUS + NTU cluster outperforms the IISc + 7IITs cluster on all three counts. The research base in the former is larger, it produces work which is uniformly of higher quality and is structurally more diverse.

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ACKNOWLEDGEMENTS. I thank the creators of the web applications, Lutz Bornmann, Division for Science and Innovation Studies, Administrative Headquarters of the Max Planck Society, Munich, Germany; Rüdiger Mutz (Professorship for Social Psychology and Research on Higher Education, ETH Zurich/MUG, Switzerland); Moritz Stefaner (Truth and Beauty Operator); and Felix de Moya Anegón, CSIC/CCHS/IPP, SCImago Group (Spain), Communication and Information Science Faculty, University of Granada, Spain for making the applications available in the public domain. I also thank Lutz Bornmann for useful discussions.

Received 19 February 2016; accepted 30 August 2016

doi: 10.18520/cs/v112/i05/1012-1015

ABCD matrix formalism to determine nonlinear refraction coefficient by Z-scan technique

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In this study, we revisit the popular method of measuring the nonlinear susceptibility of a material through Z-scan technique, introduced in 1990 by Sheik-Bahae and co-workers through a simple ray optics defined by the ABCD matrix formulation. The work therefore looks at the Z-scan measurement curves analysed through ray propagation in the medium and analysed through an aperture. The transmittance of a sample in the Z-scan technique is measured through a finite aperture in the far field, as the sample is scanned along the propagation direction (Z) of a focussed Gaussian beam. The sign and magnitude of nonlinear refractive index are easily deduced from the transmittance curve (Z-scan) using the theoretical model based on ABCD matrix formalism.

Keywords: ABCD ray matrix, linear optics, nonlinear optics, *Z*-scan technique.

ABCD matrix formalism is an efficient and widely used tool to describe the propagation of a beam through arbitrary optical systems. ABCD matrices for free propagation and for many optical components (lens, mirror, etc.) are known[1,2] and extensively used in commercial ray tracing softwares like ZEMAX, Code-V, etc. for design and analysis of complex optical systems. These matrices are also useful to determine the characteristics of paraxial optical systems, such as their effective focal length and the position of their six cardinal points. They are used to characterize the width and the wavefront curvature of an optical gaussian beam after its propagation through different optical components. The present work attempts to use the ABCD matrix formulation to describe the Z-scan technique to determine the nonlinear response of a material. There are several methods to measure nonlinear refraction including nonlinear interferometry[3,4], degenerate four-wave mixing[5], degenerate three-wave mixing[6], ellipse rotation[7], and beam distortion measurements[8] and Z-scan[9,10]. The first three methods are potentially sensitive techniques, but these require relatively complex experimental apparatus, whereas Z-scan is a simple technique to study nonlinear refraction and nonlinear absorption. It has been shown that nonlinear refraction and its sign can be obtained from a simple linear relationship between the

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observed transmittance versus sample position with respect to the focal point on the propagation axis (Z)[9].

The basics of nonlinear optics necessary for Z-scan are presented and the theoretical analysis of Z-scan measurements using ABCD matrix assuming an intensity-dependent refractive index component for analysing the transmitted beam parameters is presented. Experiments for measuring the third-order nonlinearity via Z-scan were performed on CS2 sample, comparing this data with theoretical simulation by using ABCD matrix formalism. The nonlinear refractive index (n_2) was calculated which agrees well with the literature value.

Here, we demonstrate a simple single beam method for measuring sign and magnitude of nonlinear refractive index (n_2) using ABCD matrix formalism for analysing the beam properties. Experiments were carried out with a femtosecond laser and the results show that three-photon absorption contributes to the measurements.

The dipole moments per unit volume, or polarization $\tilde{P}(t)$, of a material system depends on the strength of the applied electric field, $\tilde{E}(t)$. In the case of linear optics, the induced polarization is proportional to electric field, i.e.

$$\tilde{P}(t) = \chi^{(1)}\tilde{E}(t), \tag{1}$$

where $\chi^{(1)}$ is the linear susceptibility second rank tensor.

Under the application of intense electric fields via laser beams, the polarization, representing the electronic response of the medium can be treated as a power series in the electric field strength $\tilde{E}(t)$ given as

$$\tilde{P}(t) = \chi^{(1)}\tilde{E}(t) + \chi^{(2)}\tilde{E}^2(t) + \chi^{(3)}\tilde{E}^3(t) + \ldots. \tag{2}$$

The quantities $\chi^{(2)}$ and $\chi^{(3)}$ are second-order and third-order nonlinear optical susceptibilities respectively which are third and fourth rank tensors[11]. For centro-symmetric system, i.e. the media which show inversion symmetry, all the even order terms like $\chi^{(2)}$, $\chi^{(4)}$, $\chi^{(6)}$… become zero. For all centro-symmetric media, $\chi^{(3)}$ is the lowest surviving nonlinear term. The present experiments were performed on centro-symmetric media, to measure the real parts of the complex term $\chi^{(3)}$ through the Z-scan technique by applying the ABCD matrix formalism. The refractive index of the nonlinear medium is expressed in terms of nonlinear index n_2 (cm^2/W)[11] through

$$n = n_0 + n_2 I, \tag{3}$$

where n_0 is the linear refractive index, and $I = (cn_0/8\pi)|\tilde{E}|^2$, is the irradiance of the laser beam within the sample and n_2 is related to the nonlinear susceptibility through

$$n_2 = \frac{12\pi^2}{cn_0^2}\chi^{(3)}. \tag{4}$$

As per eq. (3), a spatially Gaussian light beam passing through a nonlinear medium will experience a varying refractive index across its cross-section, due to the radial intensity variation from beam centre to the periphery. Depending on the positive or negative sign of the nonlinear susceptibility ($\chi^{(3)}$) or nonlinear refractive index (n_2), one observes self-focussing (positive n_2) and self-defocussing (negative n_2) of the incident light. As the third-order nonlinear susceptibility is generally complex in nature, its real part contributes to refractive index change and the imaginary part contributes to the absorption co-efficient[11].

The popular Z-scan technique is experimentally a simple and sensitive method for measuring both nonlinear refraction and nonlinear absorption, which was introduced by Sheik-Bahae *et al.*[9] in 1990. The following explanation gives a qualitative idea on how such a trace (Z-scan) is related to nonlinear refraction of the sample. Assume, for instance, a sample with negative nonlinear refractive index and its thickness is smaller than the Rayleigh length of the focussed beam (a thin medium). This can be regarded as a thin lens (negative or concave lens) of variable focal length at different positions of the sample on the Z-axis. Starting the scan from a distance far away from the focus (negative Z), where the beam irradiance is low and one expects negligible nonlinear refraction (Figure 1), the transmittance (D_2/D_1), the ratio of signal detected by detector D_2 to the signal detected by reference detector (D_1) remains relatively constant.

As the sample is brought closer to the focus, the beam irradiance increases, leading to self-lensing in the sample due to its nonlinearity. A negative self-lensing prior to focus will try to collimate the beam, causing a beam narrowing near the aperture resulting in an increase in the measured transmittance. As the scan in Z continues and the sample passes the focal plane to the right (positive Z), the same negative lensing effect increases the beam divergence, leading to beam broadening at the aperture, and thus a decrease in transmittance through the aperture. It is also easy to realize that there will be no change at $Z = 0$, as it is analogous to placing a thin lens at the focus, resulting in a minimal change to the far field pattern of the beam. The Z-scan is completed as the sample is moved far away from focus (positive Z) such that the transmittance becomes linear, which happens when the irradiance is again low[9].

Let us derive the equation for the beam waist at the detector as a function of the sample position (Z), using the ABCD matrices for convex lens, free space and the sample. Consider the geometry given in Figure 1. It is easy to formulate ABCD matrix formalism for analysing the Z-scan data. From Figure 2, w_0 is the minimum spot size, z_0 the Rayleigh range, r_1 the input beam waist, f_1 the focal length of the convex lens, f_2 the induced focal length for the sample due to incident Gaussian beam, r_2 the beam waist at the aperture, x the distance between lens and the

sample and y is the distance between the sample and aperture. $D = (x + y)$ is the distance between lens and aperture, and w_z is the beam waist at different positions (Z) given by[12]

$$w_z = w_0\sqrt{1+\left(\frac{z}{z_0}\right)^2}.$$

When the Gaussian beam propagates through the sample, the medium acts as a lens, whose focal length depends on the intensity of beam at that point. By scanning the sample from $-Z$ to $+Z$, the change in beam waist at aperture (r_2) is observed.

First we relate beam waist (r_2) as a function of Z using ABCD matrices for all the optical components and then do a numerical integration across the beam to arrive at the nonlinear transmittance (T) as a function of the sample position (Z).

From Figure 2, the ABCD matrices for convex lens, free space between lens and sample, sample and free space between sample and aperture are respectively given by

$$\begin{pmatrix} 1 & 0 \\ -\frac{1}{f_1} & 1 \end{pmatrix}, \begin{pmatrix} 1 & x \\ 0 & 1 \end{pmatrix}, \begin{pmatrix} 1 & 0 \\ -\frac{1}{f_2} & 1 \end{pmatrix} \text{ and } \begin{pmatrix} 1 & y \\ 0 & 1 \end{pmatrix}.$$

The resultant ABCD matrix is therefore a multiplication of these four matrices, from which we arrive at the beam waist at the aperture (r_2) as given by

$$r_2 = \frac{1}{f_1 f_2}[-r_1 f_2 y + xyr_1 - r_1 f_1 y - xr_1 f_2 + r_1 f_1 f_2]. \quad (6)$$

The nonlinear optical element is the sample, whose focal length f_2 is a function of the sample position (Z)[13] because of the varying intensity produced by the first lens.

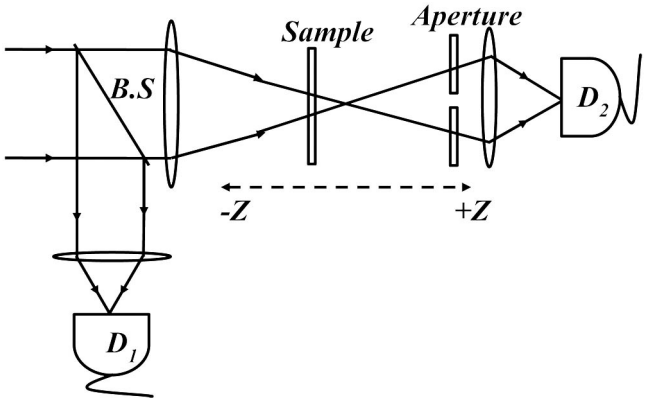

Figure 1. Simple Z-scan apparatus for measuring the nonlinear transmittance as a function of sample position z.

We estimate the focal length f_2 of the sample using the lens maker's formula. Figure 3 shows the calculation of light-induced radius of curvature for the sample (R). From Figure 3, $R^2 + b^2 - 2Rh + h^2 + R^2$ (since $h << b$),

$$R = \frac{b^2}{2h}, \quad (7)$$

where

$$2h = tn_2 I_{00}, \quad (8)$$

where t = 1 mm is the thickness of the sample used in the experiment, and $2h$ is the apparent thickness variation of the medium (Figure 3) at the centre of the Gaussian profile by assuming that the refractive index does not vary, but only the thickness varies. This enables us to think of the medium as a convex lens with an increase in thickness of $2h$ at the centre of the Gaussian beam for a positive nonlinear medium and a reduction in the thickness by $2h$ for a negative nonlinear medium.

As the area covered by the beam in the sample leads to the formation of intensity dependent lens, b is equal to w_z. The intensity along z is

$$I_{00} = \frac{P}{\pi W_z^2}, \quad (9)$$

where P is equal to the total power incident on the sample. It is assumed that the absorbance in the sample is

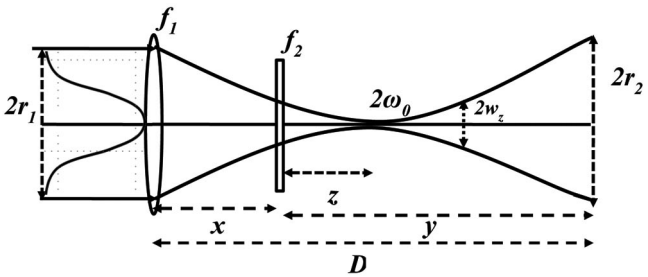

Figure 2. Schematic diagram for gaussian beam waist using ABCD matrix.

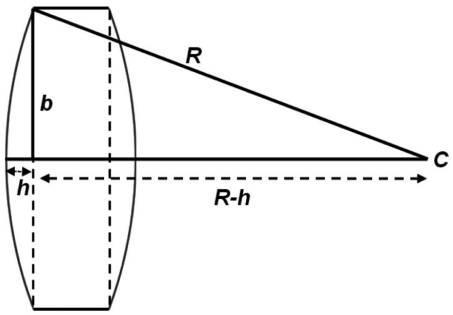

Figure 3. Calculation of the radius of curvature of the sample.

small. Equation (9) shows that the intensity (I_{00}) is a function of beam waist along z. So the intensity and area of the beam changes, while the total power remains constant[12]. From the well-known formula for a lens, we have

$$\frac{1}{f_2} = (n-1)\left(\frac{1}{R_1} - \frac{1}{R_2}\right), \tag{10}$$

where n is the refractive index of the nonlinear medium which is given by $n = n_0 + n_2 I$ and R_1, R_2 are the radii of curvature of the two surfaces of the induced lens. If we assume negligible absorption, $R_1 \approx -R_2 = R$. From the eq. (10), we obtain

$$f_2 = \frac{R}{2(n-1)}. \tag{11}$$

Substituting f_2, in eq. (6), we have r_2 as a function of z. The variation of r_2 with sample position Z is shown in Figure 4 *a* and *b* for negative and positive nonlinear refractive indices respectively.

Figure 4 *a* illustrates the case of $n_2 < 0$, where the sample is scanned from $-Z$, the beam irradiance increases slowly and the sample acts as a concave lens making the equivalent focal point shift towards $+Z$ direction. As a result, the beam waist decreases at the aperture, so that we have a valley first. When the sample is in $+Z$ position, the beam again diverges at the aperture, so that we have a peak for $z > 0$. For positive nonlinearity ($n_2 > 0$), an exactly opposite process happens, which is shown in Figure 4 *b*.

Once we have r_2 as a function of z, by numerical integration of the Gaussian function over aperture area, we can obtain power through the aperture. Also by integrating the same Gaussian function over the total area (the area covered by beam at aperture), we obtain the total power. Division of the power through aperture (P_a) with the total transmittance power (P_t) gives the nonlinear transmittance (T). We now discuss the numerical integration of Gaussian function over the aperture. The Gaussian function is

$$F = F_0 \mathrm{e}^{-(r/r_2)^2}, \tag{12}$$

where F_0 is the amplitude of Gaussian beam[12] which is a function of z. Now

$$P_a = \int_0^a F \times 2\pi r \, \mathrm{d}r, \tag{13}$$

where r varies from 0 to a (a is the aperture size). Similarly we have,

$$P_t = \int_0^{r_2} F \times 2\pi r \, \mathrm{d}r,$$

Nonlinear transmittance

$$T = \frac{\text{Power through the aperture } (P_a)}{\text{Total power } (P_t)}. \tag{14}$$

Figure 5 *a* and *b* show the variation of nonlinear transmittance (T) with the sample position (Z) for both negative

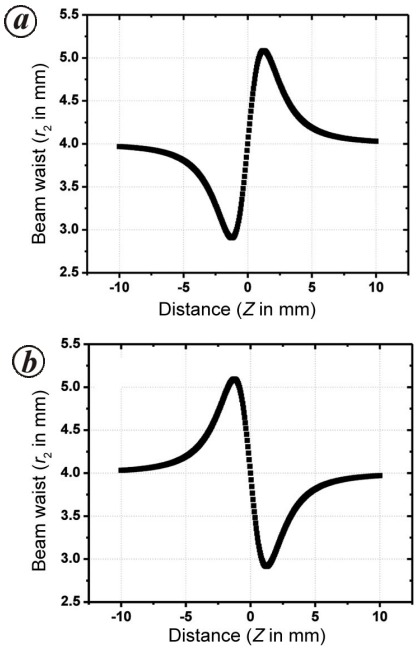

Figure 4. Graph represents variation of beam waist at the aperture with the sample position (z) for (***a***) $n_2 < 0$ and (***b***) $n_2 > 0$. The results obtained using ABCD matrix formalism.

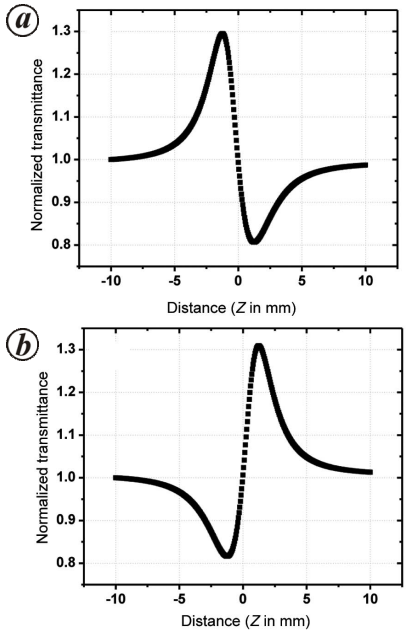

Figure 5. Graph represents nonlinear transmittance versus sample position using ABCD matrix formalism in two cases (***a***) for $n_2 < 0$ and (***b***) for $n_2 > 0$.

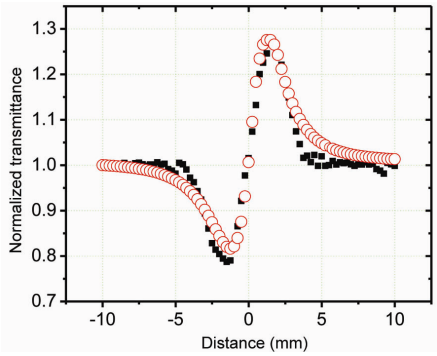

Figure 6. Closed aperture signature for CS_2 material by comparing experimental Z-scan and ABCD matrix formalism dates. *Y*-axis represents normalized nonlinear transmittance.

and positive n_2 values respectively[9]. In the case of $n_2 < 0$, the beam irradiance increases at the aperture, which causes an increase in power through aperture and hence we have a peak first in Figure 5 *a*. When the sample is brought to $+Z$, the irradiance decreases at aperture and hence we have a valley for $+Z$. As a result, we obtain a peak followed by a valley which is the negative nonlinearity as shown in Figure 5 *a*. An exactly opposite process happens for $n_2 > 0$ as shown in Figure 5 *b*.

CS_2 is often used as a reference while measuring the nonlinearities of any new material[9,11]. When measurements are carried out with an ultra-short femtosecond (fs) laser pulse, slow time scale phenomena such as thermal process are no longer significant and can be ignored. Particularly, in CS_2 the molecular re-orientational Kerr effect becomes the dominant mechanism for nonlinear refraction. Using femtosecond pulses having a repetition rate 1 kHz, wavelength ~800 nm, and pulse width of ~110 fs, the nonlinear refractive index of CS_2 was measured.

The measurement was carried out in a solution with 1 mm thickness cuvette and using fs laser pulses focussed with a beam waist $2w_0$ of 40 μm from a Ti-sapphire laser. Femtosecond laser pulse has an intensity of $(I_0) = 3.98 \times 10^{11}$ W/cm^2 at its focus, which was achieved by using a convex lens of focal length 120 mm, incident beam radius $(2r_1)$ of 3 mm and Rayleigh range (Z_0) of 1.57 mm.

The collecting aperture in front of the detector is kept at 480 mm from the focussing convex lens, and its size is approximately one fourth of r_1. With the help of translation stage monitored by stepper motor, the sample is scanned along Z direction, and the transmitted light was collected through an aperture kept in front of the detector. Experimental data in Figure 6 shows that the valley-peak configuration of CS_2 infers the positive sign of n_2. This positive nonlinearity is due to Kerr effect in CS_2. The black square dotted and red round dotted data points indicate the experimental and theoretical results respectively, in Figure 6. The theoretical data points with respect to sample position were obtained from eq. (14) by ABCD matrix formulation method. The best fit for the experimental data using the theoretical equation yields the nonlinear refractive index $(n_2) = 0.91 \times 10^{-19}$ m^2/W, which is very close to literature value[14]. This work proves that the evaluation of third order nonlinear refractive index can be achieved accurately using ABCD formulation.

We used ABCD matrix formalism of the Z-scan experimental configuration to obtain nonlinear refractive index of a standard sample such as CS_2. The sign and magnitude of nonlinear refractive index of CS_2 was measured and compared with literature value, which matches well with estimated value. We therefore conclude that the ABCD matrix formalism can be effectively used to measure nonlinearities of any nonlinear material. We believe that this ABCD matrix formulation has mathematical ease, especially for simulating Gaussian beam propagation through nonlinear medium.

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ACKNOWLEDGEMENT. We acknowledge financial support by DST through ITPAR project.

Received 19 March 2015; revised accepted 20 September 2016

doi: 10.18520/cs/v112/i05/1015-1019