

Z-SCAN: A SIMPLE AND SENSITIVE TECHNIQUE FOR NONLINEAR REFRACTION MEASUREMENTS

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ABSTRACT

We describe a sensitive technique for measuring nonlinear refraction in a variety of materials that offers simplicity, sensitivity and speed. The transmittance of a sample is measured through a finite aperture in the *far-field* as the sample is moved along the propagation path (z) of a focused Gaussian beam. The sign and magnitude of the nonlinearity is easily deduced from such a transmittance curve (Z-scan). Employing this technique a sensitivity of better than $\lambda/300$ wavefront distortion is achieved in n_2 measurements of BaF_2 using picosecond frequency doubled Nd:YAG laser pulses.

1. INTRODUCTION

We are currently developing a single beam method, which we refer to as a Z-scan, for measuring the sign and magnitude of the nonlinear refractive index n_2 . [1] In practice we have found that this method has a sensitivity comparable to interferometric methods. Here we describe this method in detail and demonstrate how it can be applied and analyzed for a variety of materials. We also present a simple method to minimize parasitic effects due to the presence of linear sample inhomogeneities.

Previous measurements of nonlinear refraction have used a variety of techniques including nonlinear interferometry [2], [3], degenerate four-wave mixing [4], nearly-degenerate three-wave mixing [5], ellipse rotation [6], beam distortion measurements [7], [8], and our recently reported Z-scan technique. The first three methods, namely nonlinear interferometry and wave mixing are potentially sensitive techniques but require a relatively complex experimental apparatus. Beam distortion measurements, on the other hand, are relatively insensitive and require detailed wave propagation analysis. The Z-scan technique is based on the principles of spatial beam distortion but offers simplicity as well as very high sensitivity.

We will describe this simple technique in Section II. Theoretical analyses of Z-scan measurements are given in Section III for a "thin" nonlinear medium. It will be shown that for many practical cases, nonlinear refraction and its sign can be obtained from a simple linear relationship between the observed transmittance changes and the induced phase distortion without the need for performing detailed calculations. In Section IV we present measurements of nonlinear refraction in a number of materials such as CS_2 , and transparent dielectrics at wavelengths of 532 nm, 1.06 μm and 10.6 μm . In CS_2 at 10 μm , for example, both thermo-optical and reorientational Kerr effects were identified using nanosecond and picosecond pulses respectively. We also describe how effects of linear sample inhomogeneities (eg. bulk index variations) can be effectively removed from the experimental data.

2. THE Z-SCAN TECHNIQUE

Using a single Gaussian laser beam in a tight focus geometry, as depicted in Fig. 1, we measure the transmittance of a nonlinear medium through a finite aperture *in the far field* as a function of the sample position z measured with respect to the focal plane. The following example will qualitatively elucidate how such a trace (Z-scan) is related to the nonlinear refraction of the sample. Assume, for instance, a material

with a negative nonlinear refractive index and a thickness smaller than the diffraction length of the focused beam (a thin medium). This can be regarded as a thin lens of variable focal length. Starting the scan from a distance far away from the focus (negative z) the beam irradiance is low and negligible nonlinear refraction occurs; hence, the transmittance (D_2/D_1 in Fig. 1) remains relatively constant. As the sample is brought closer to focus, the beam irradiance increases leading to self-lensing in the sample. A negative self-lensing prior to focus will tend to collimate the beam, causing a beam narrowing at the aperture which results 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 self-defocusing increases the beam divergence leading to beam broadening at the aperture and, thus, a decrease in transmittance. This suggests that there is a null as the sample crosses the focal plane. This is analogous to placing a thin lens at or near the focus, resulting in a minimal change of the far field pattern of the beam. The Z-scan is completed as the sample is moved away from focus (positive z) such that the transmittance becomes linear since the irradiance is again low. Induced beam broadening and narrowing of this type have been previously observed and explained during nonlinear refraction measurements of some semiconductors.[9],[10] A similar technique was also previously used to measure thermally induced beam distortion of chemicals in solvents.[11]

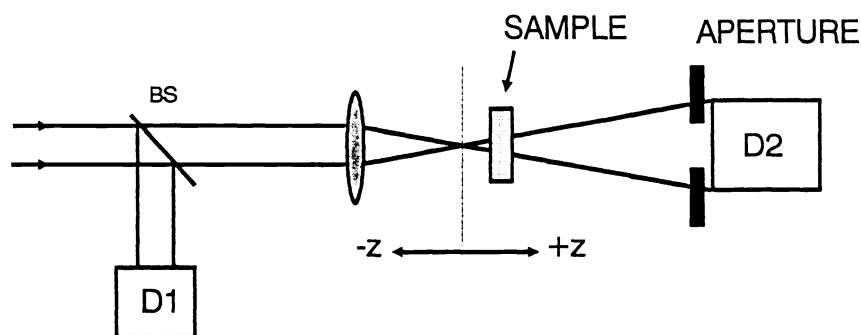


Fig.1 The Z-scan experimental apparatus in which the ratio $D2/D1$ is recorded as a function of the sample position z .

A pre-focal transmittance maximum (peak) followed by a post-focal transmittance minimum (valley) is, therefore, the Z-scan signature of a negative refractive nonlinearity. Positive nonlinear refraction, following the same analogy, gives rise to an opposite valley-peak configuration. It is an extremely useful feature of the Z-scan method that the sign of the nonlinear index is immediately obvious from the data, and as we will show in the following section the magnitude can also be easily estimated using a simple analysis for a thin medium.

In the above picture describing the Z-scan, one must bear in mind that a purely refractive nonlinearity was considered assuming that no absorptive nonlinearities (such as multiphoton or saturation of absorption) are present. Qualitatively, multiphoton absorption suppresses the peak and enhances the valley, while saturation produces the opposite effect. The sensitivity to nonlinear refraction is entirely due to the aperture, and removal of the aperture completely eliminates the effect. However, in this case the Z-scan will still be sensitive to nonlinear absorption. Nonlinear absorption coefficients could be extracted from such "open" aperture experiments.

3. THEORY

Much work has been done in investigating the propagation of intense laser beams inside a nonlinear material and the ensuing self-refraction [12], [13]. Considering the geometry given in Fig. 1, we will formulate and discuss a simple method for analyzing the Z-scan data based on modifications of existing theories.

In general, various order nonlinearities can be considered; however, for simplicity, we first examine only a cubic nonlinearity where the index of refraction n is expressed in terms of nonlinear indices $n_2(\text{esu})$ or $\gamma(\text{m}^2/\text{W})$ through:

$$n = n_0 + \frac{n_2}{2} |E|^2 = n_0 + \gamma I \quad (1)$$

where n_0 is the linear index of refraction, E is the peak electric field (cgs), and I denotes the irradiance (MKS) of the laser beam within the sample. [n_2 and γ are related through the conversion formula, $n_2(\text{esu}) = (cn_0/40\pi)\gamma(\text{m}^2/\text{W})$, where c (m/sec) is the speed of light in vacuum]. Assuming a TEM₀₀ Gaussian beam of beam waist radius w_0 traveling in the + z direction, we can write E as:

$$E(z,r,t) = E_0(t) \frac{w_0}{w(z)} \exp\left[-\frac{r^2}{w^2(z)} - \frac{ikr^2}{2R(z)}\right] e^{-i\phi(z,t)}, \quad (2)$$

where $w^2(z) = w_0^2(1+z^2/z_0^2)$ is the beam radius, $R(z) = z(1+z_0^2/z^2)$ is the radius of curvature of the wavefront at z , $z_0 = kw_0^2/2$ is the diffraction length of the beam, $k = 2\pi/\lambda$ is the wave vector and λ is the laser wavelength, all in free space. $E_0(t)$ denotes the radiation electric field at the focus and contains the temporal envelope of the laser pulse. The $e^{-i\phi(z,t)}$ term contains all the radially uniform phase variations. As we are only concerned with calculating the radial phase variations $\Delta\phi(r)$, the slowly varying envelope approximation (SVEA) applies, and all other phase changes that are uniform in r are ignored.

If the sample length is small enough that changes in the beam diameter within the sample due to either diffraction or nonlinear refraction can be neglected, the medium is regarded as "thin", in which case the self-refraction process is referred to as "external self-action". Such an assumption simplifies the problem considerably, and the amplitude \sqrt{I} and phase ϕ of the electric field as a function of z' are now governed in the SVEA by a pair of simple equations:

$$\frac{d\Delta\phi}{dz'} = \Delta n(I) k, \quad (3)$$

and

$$\frac{dI}{dz'} = -\alpha I, \quad (4)$$

where z' is the propagation depth in the sample and α is the linear absorption coefficient. Note that z' should not be confused with the sample position z . In the case of a cubic nonlinearity, Eqns. 3 and 4 are solved to give the phase shift $\Delta\phi$ at the exit surface of the sample, which simply follows the radial variation of the incident irradiance at a given position of the sample z . Thus,

$$\Delta\phi(z,r,t) = \Delta\phi_0(z,t) \exp\left[-\frac{2r^2}{w^2(z)}\right], \quad (5-a)$$

with

$$\Delta\phi_0(z,t) = \frac{\Delta\Phi_0(t)}{1+z^2/z_0^2} . \quad (5-b)$$

$\Delta\Phi_0(t)$, the on-axis phase shift at the focus, is defined as,

$$\Delta\Phi_0(t) = k\Delta n_0(t) \frac{1-e^{-\alpha L}}{\alpha} , \quad (6)$$

where L is the sample length, and $\Delta n_0 = \gamma I_0(t)$ with $I_0(t)$ being the on-axis irradiance at focus (ie. $z=0$). Again we take $I_0(t)$ as the irradiance within the sample to account for Fresnel reflection losses.

The complex electric field after the sample, E' , now contains the nonlinear phase distortion,

$$E' = E(z,r,t) e^{-\alpha L/2} e^{i\Delta\phi(z,r,t)} . \quad (7)$$

By virtue of Huygen's principle one can obtain the far field pattern of the beam at the aperture plane through a zeroth order Hankel transformation of E' . [14] We will follow a more convenient treatment applicable to Gaussian input beams which we refer to as the "Gaussian Decomposition" (GD) method given by Weaire et. al. [15], in which they decompose the complex electric field at the exit plane of the sample into a summation of Gaussian beams through a Taylor series expansion of the nonlinear phase term $e^{i\Delta\phi(z,r,t)}$ in Eq. 7. That is,

$$e^{i\Delta\phi(z,r,t)} = \sum_{m=0}^{\infty} \frac{[i\Delta\phi_0(z,t)]^m}{m!} e^{-2mr^2/w^2(z)} . \quad (8)$$

Each Gaussian beam can now be simply propagated to the aperture plane where they will be resummed to reconstruct the beam. When including the initial beam curvature for the focused beam, we derive the resultant electric field pattern at the aperture as:

$$E_a(r,t) = E(z,r=0,t) e^{-\alpha L/2} \sum_{m=0}^{\infty} \frac{(i\Delta\phi_0(t))^m}{m!} \left(g^2 + \frac{d^2}{d_m^2} \right)^{-\frac{1}{2}} \exp \left[-\frac{r^2}{w_m^2} - \frac{ikr^2}{2R_m} + i\theta_m \right] , \quad (9)$$

where d is the propagation distance in free space from the sample to the aperture plane, and $g=1+d/R$, $R=R(z)$ being the beam radius of curvature at the sample. As long as the far field condition is met, d can be considered independent of the sample position z resulting in symmetric Z-scans. The remaining parameters in Eq. 9 are expressed as:

$$w_{m0}^2 = \frac{w^2(z)}{2m+1} , \quad d_m = \frac{kw_{m0}^2}{2} , \quad w_m^2 = w_{m0}^2 \left[g^2 + \frac{d^2}{d_m^2} \right] ,$$

$$R_m = d \left[1 - \frac{g}{g^2 + d^2/d_m^2} \right]^{-1} , \quad \text{and } \theta_m = \tan^{-1} \left[\frac{d/d_m}{g} \right] .$$

The expression given by Eq. 9 is a general case of that derived in Ref. [15] where they considered a collimated beam ($R=\infty$) for which $g=1$. We find that this GD method is very useful for the small phase distortions detected with the Z-scan method since only a few terms of the sum in Eq. 9 are needed. The method is also easily extended to higher order nonlinearities.

The transmitted power through the aperture is obtained by spatially integrating $E_a(r,t)$ up to the aperture radius r_a , giving,

$$P_T(\Delta\Phi_0(t)) = \frac{c\epsilon_0 n_0}{2} \int_0^{r_a} |E_a(r,t)|^2 r dr . \quad (10)$$

Including the pulse temporal variation, the normalized Z-scan transmittance $T(z)$ can be calculated as:

$$T(z) = \frac{\int_{-\infty}^{\infty} P_T(\Delta\Phi_0(t)) dt}{S \int_{-\infty}^{\infty} P_i(t) dt} , \quad (11)$$

where $P_i(t) = \pi w_0^2 I_0(t)/2$ is the instantaneous input power (within the sample) and S is the aperture transmittance in the linear regime.

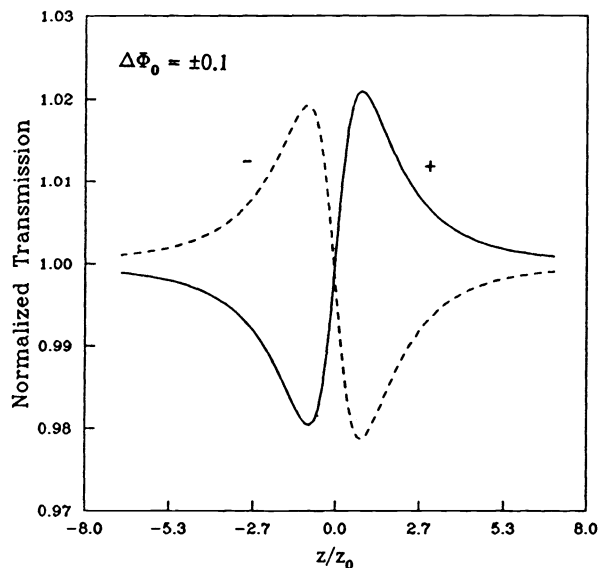


Fig.2 Calculated Z-scan transmittance curves for a cubic nonlinearity with either polarity and a small aperture ($S=0.01$).

We first consider an instantaneous nonlinearity and a temporally square pulse to illustrate the general features of the Z-scan. This is equivalent to assuming cw radiation and the nonlinearity has reached the steady state. The normalized transmittance, $T(z)$, in the far field, is shown in Fig. 2 for $\Delta\Phi_0 = \pm 0.1$ and a

small aperture ($S=0.01$). They exhibit the expected features, namely a valley-peak (v-p) for the positive nonlinearity and a peak-valley (p-v) for the negative one. For a given $\Delta\Phi_0$, the magnitude and shape of $T(z)$ do not depend on the wavelength or geometry as long as the far field condition for the aperture plane is satisfied. The aperture size S , however, is an important parameter since a large aperture reduces the variations in $T(z)$. This reduction is more prominent in the peak where beam narrowing occurs and can result in a peak transmittance which cannot exceed $(1-S)$. Needless to say, for very large aperture or no aperture ($S=1$), the effect vanishes and $T(z) = 1$ for all z and $\Delta\Phi_0$. For small $|\Delta\Phi_0|$, the peak and valley occur at the same distance with respect to focus, and for a cubic nonlinearity this distance is found to be $\approx 0.85z_0$. With larger phase distortions ($|\Delta\Phi_0| > 1$) this symmetry no longer holds and both peak and valley move toward $\pm z$ for the corresponding sign of nonlinearity ($\pm\Delta\Phi_0$) such that their separation remains relatively constant given by,

$$\Delta Z_{p-v} \approx 1.7z_0 . \quad (12)$$

We can define an easily measurable quantity ΔT_{p-v} as the difference between the normalized peak and valley transmittance: $T_p - T_v$. The variation of this quantity as a function of $|\Delta\Phi_0|$, as calculated for various aperture sizes is illustrated in Fig. 3. These curves exhibit some useful features. First, for a given order of nonlinearity, they can be considered universal. In other words, they are independent of the laser wavelength, geometry (as long as the far field condition is met) and the sign of nonlinearity. Second, for all aperture sizes, the variation of ΔT_{p-v} is found to be linearly dependent on $|\Delta\Phi_0|$. Particularly for on axis ($S \approx 0$) we find,

$$\Delta T_{p-v} \approx 0.405 |\Delta\Phi_0| \quad \text{for} \quad |\Delta\Phi_0| \leq \pi , \quad (13-a)$$

to be accurate to within 0.5 percent. As shown in Fig. 3, for larger apertures, the linear coefficient 0.405 decreases such that with $S=0.5$ it becomes 0.34 and at $S=0.7$ it reduces to 0.29. Based on a numerical fitting, the following relationship can be used to include such variations within a $\pm 2\%$ accuracy;

$$\Delta T_{p-v} \approx 0.405(1-S)^{0.25} |\Delta\Phi_0| \quad \text{for} \quad |\Delta\Phi_0| \leq \pi . \quad (13-b)$$

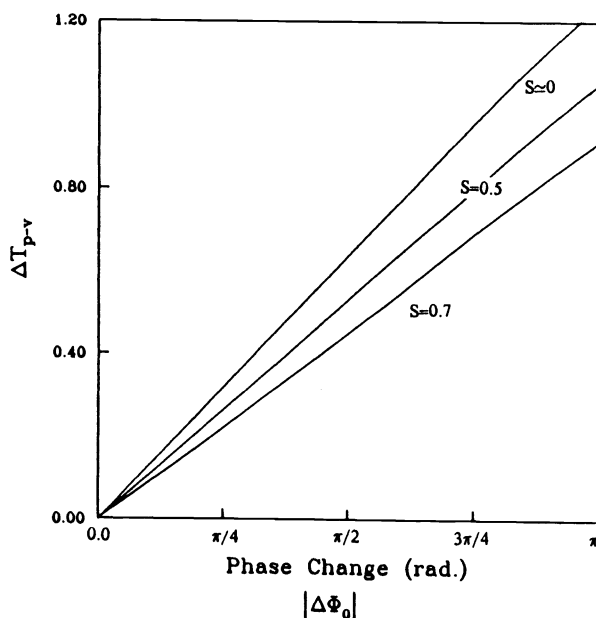


Fig.3 Calculated ΔT_{p-v} as a function of the phase shift at the focus ($\Delta\Phi_0$). The sensitivity, as indicated by the slope of the curves, decreases slowly for larger aperture sizes ($S > 0$).

The implications of Eqns. 13-a and 13-b are quite promising in that they can be used to readily estimate the nonlinear index (n_2) with good accuracy after a Z-scan is performed. What is most intriguing about these expressions is that they reveal the highly sensitive nature of the Z-scan technique. For example, if our experimental apparatus and data acquisition systems are capable of resolving transmission changes ΔT_{p-v} of $\simeq 1\%$, we will be able to measure phase changes corresponding to less than $\lambda/250$ wavefront distortion. Achieving such sensitivity, however, requires relatively good optical quality of the sample under study. We describe in the experimental section IV a means to minimize problems arising from poor optical quality samples.

We can now easily extend the steady state cw results to include transient effects along with pulsed radiation by using the time averaged index change $\langle \Delta n_0(t) \rangle$ where,

$$\langle \Delta n_0(t) \rangle = \frac{\int_{-\infty}^{\infty} \Delta n_0(t) I_0(t) dt}{\int_{-\infty}^{\infty} I_0(t) dt}, \quad (14)$$

The time averaged $\langle \Delta \Phi_0(t) \rangle$ is related to $\langle \Delta n_0(t) \rangle$ through Eq. 6. With a nonlinearity having instantaneous response and decay times relative to the pulsewidth of the laser, one obtains for a temporally Gaussian pulse:

$$\langle \Delta n_0(t) \rangle = \Delta n_0 / \sqrt{2}, \quad (15)$$

where Δn_0 now represents the peak-on-axis index change at the focus. For a cumulative nonlinearity having a decay time much longer than the pulsewidth (eg. thermal), the instantaneous index change is given by the following integral:

$$\Delta n_0(t) = A \int_{-\infty}^t I_0(t') dt', \quad (16)$$

where A a constant which depends on the nature of the nonlinearity. If we substitute Eq. 16 into Eq. 14 we obtain a fluence averaging factor of 1/2. That is,

$$\langle \Delta n_0(t) \rangle = \frac{1}{2} A F, \quad (17)$$

where F is the pulse fluence at focus within the sample. Interestingly, the factor of 1/2 is independent of the temporal pulse shape.

4. EXPERIMENTAL RESULTS

We examined the nonlinear refraction of a number of materials using the Z-scan technique. Fig. 4 shows a Z-scan of a 1 mm thick cuvette with NaCl windows filled with CS_2 using 300 ns TEA CO_2 laser pulses having an energy of 0.85 mJ. The peak-valley configuration of this Z-scan is indicative of a negative (self-defocusing) nonlinearity. The solid line in Fig. 4 is the calculated result using $\langle \Delta \Phi_0 \rangle = -0.6$ which gives an index change of $\langle \Delta n_0 \rangle \simeq -1 \times 10^{-3}$. As mentioned earlier such detailed theoretical fitting is not necessary

for obtaining $\langle \Delta n_0 \rangle$ (only ΔT_{p-v} is needed). The defocusing effect shown in Fig. 4 is attributed to a thermal nonlinearity resulting from linear absorption of CS_2 ($\alpha \approx 0.22 \text{ cm}^{-1}$ at $10.6 \mu\text{m}$). The risetime of a thermal lens in a liquid is determined by the acoustic transit time, $\tau = w_0/v_s$, where v_s is the velocity of sound in the liquid [17]. For CS_2 with $v_s \approx 1.5 \times 10^5 \text{ cm/sec}$ and having $w_0 \approx 60 \mu\text{m}$, we obtain a risetime of $\approx 40 \text{ ns}$ which is almost an order of magnitude smaller than the TEA laser pulsewidth. Furthermore, the relaxation of the thermal lens, governed by thermal diffusion, is of the order of 100 ms.[17] Therefore, we regard the nonuniform heating caused by the 300 ns pulses as quasi-steady state, in which case, from Eq. 17, the average on-axis nonlinear index change at focus can be determined in terms of the thermo-optic coefficient, dn/dT , as:

$$\langle \Delta n_0 \rangle \approx \frac{dn}{dT} \frac{F_0 \alpha}{2\rho C_v}, \quad (18)$$

where F_0 is the fluence, ρ is the density, C_v is the specific heat and $1/2$ denotes the fluence averaging factor. With the known value of $\rho C_v \approx 1.3 \text{ J/}^\circ\text{Kcm}^3$ for CS_2 , we deduce $dn/dT \approx -(8.3 \pm 1.0) \times 10^{-4} \text{ }^\circ\text{C}^{-1}$ which is in good agreement with the reported value of $-8 \times 10^{-4} \text{ }^\circ\text{C}^{-1}$. [16]

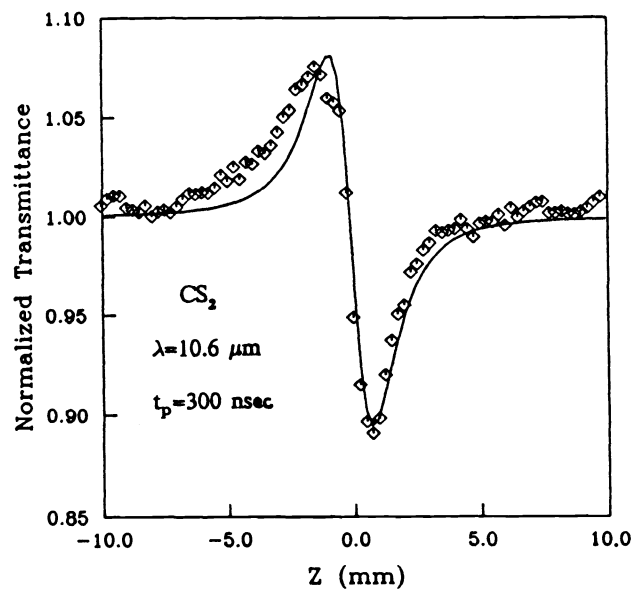


Fig.4 Measured Z-scan of a 1mm thick CS_2 cell using 300 ns pulses at $\lambda=10.6 \mu\text{m}$ indicating thermal self-defocusing. The solid line is the calculated result with $\Delta\Phi_0=-0.6$ and 60% aperture ($S=0.6$).

With ultrashort pulses, nonlocal nonlinearities such as thermal or electrostriction are no longer significant. Particularly, in CS_2 , the molecular reorientational Kerr effect becomes the dominant mechanism for nonlinear refraction. CS_2 is frequently used as a standard reference nonlinear material.[18,19] We have used picosecond pulses at $10.6 \mu\text{m}$, $1.06 \mu\text{m}$ and at $0.53 \mu\text{m}$ to measure n_2 in CS_2 . We obtain the same value of n_2 , within errors, at all three wavelengths, $(1.5 \pm 0.6) \times 10^{-11} \text{ esu}$ at $10.6 \mu\text{m}$, $(1.3 \pm 0.3) \times 10^{-11} \text{ esu}$ at $1.06 \mu\text{m}$ and $(1.2 \pm 0.2) \times 10^{-11} \text{ esu}$ at $0.53 \mu\text{m}$. The external self-focusing arising from the Kerr effect in CS_2 is shown in Fig. 5, where a Z-scan of a 1mm cell using 27 ps (FWHM) pulses focused to a beam waist w_0 of $26 \mu\text{m}$ from a frequency doubled Nd:YAG laser is illustrated. Its valley-peak configuration indicates the positive sign of n_2 . With $\Delta T_{p-v} = 0.24$, and using Eq. 13-b with a 40 percent aperture ($S = 0.4$), one readily obtains a $\langle \Delta n_0 \rangle = 5.6 \times 10^{-5}$. Using the peak irradiance of 2.6 GW/cm^2 , this value of $\langle \Delta n_0 \rangle$ corresponds to an $n_2 \approx (1.2 \pm 0.2) \times 10^{-11} \text{ esu}$. The main source of uncertainty in the value of n_2 is the absolute measurement of the irradiance. A plot of ΔT_{p-v} versus peak laser irradiance as measured from

various Z-scans on the same CS₂ cell is shown in Fig. 6. The linear behavior of this plot follows Eq. 13 as derived for a cubic nonlinearity.

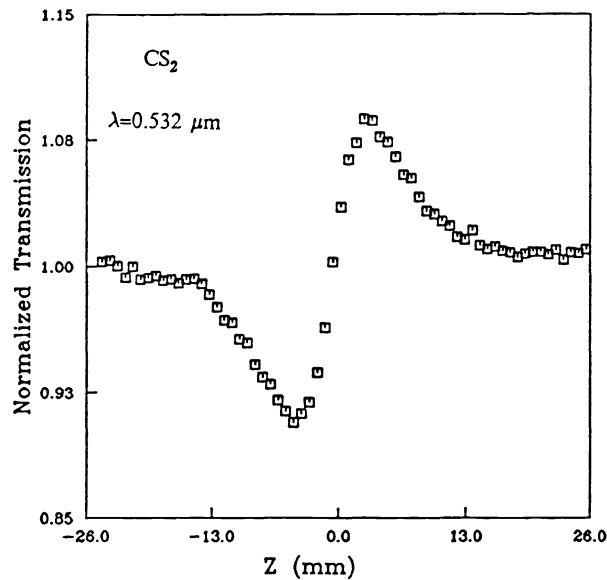


Fig.5 Measured Z-scan of a 1mm thick CS₂ cell using 27 ps pulses at $\lambda=532$ nm. It depicts the self-focusing effect due to the reorientational Kerr effect.

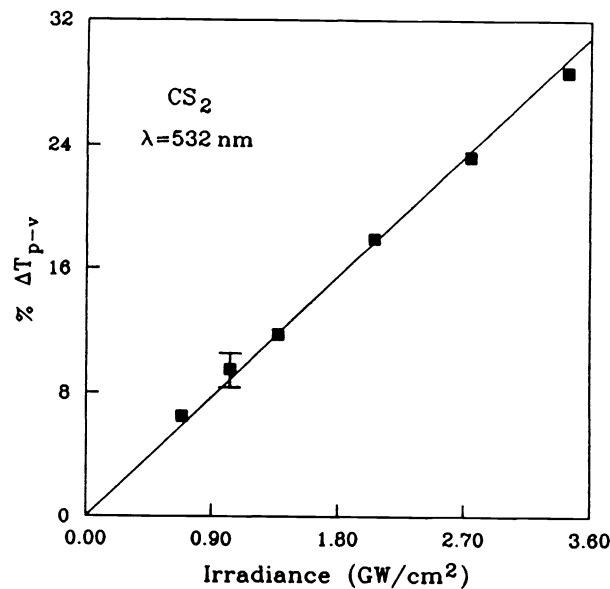


Fig.6 T_{p-v} in percent as a function of the peak irradiance from the Z-scan data of CS₂ at 532 nm, indicative of the reorientational Kerr effect.

Transparent dielectric window materials have relatively small nonlinear indices. Recently, Adair et. al. [21] have performed a careful study of the nonlinear index of refraction of a large number of such materials in a nearly degenerate-three-wave-mixing scheme at $\lambda \approx 1.06$ μm . Using the Z-scan technique, we examined some of these materials at 532 nm. For example, the result for BaF₂ (2.4mm thick) is shown in Fig.7, using the same beam parameters as for CS₂. This Z-scan was obtained by purposely lowering the pulse energy to

2 μJ in order to observe the resolution and the sensitivity of this measurement. With a $\Delta T_{p-v} \simeq 0.035$, this Z-scan corresponds to a $\lambda/75$ induced phase distortion. For a unity signal-to-noise-ratio for our particular laser system, it is seen from Fig. 7 that the sensitivity to phase distortion is better than $\lambda/300$. For laser systems having better amplitude and pulsedwidth stability, the sensitivity should be correspondingly increased.

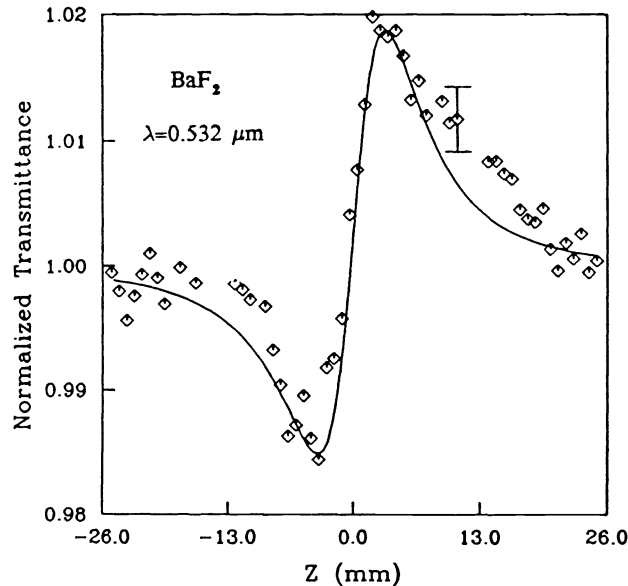


Fig.7 Measured Z-scan of a 2.4 mm thick BaF_2 sample using 20 ps pulses at $\lambda=532$ nm indicating the self-focusing due to the electronic Kerr effect. The solid line is the calculated result with $\Delta\Phi_0=0.085$ corresponding to $\simeq\lambda/75$ total phase distortion. The error bar shown corresponds to approximately $\lambda/480$ induced phase distortion.

Aside from the statistical fluctuations of the laser irradiance, surface imperfections or wedge in the sample may lead to systematic transmittance changes with z that could mask the effect of nonlinear refraction. We found, however, that such "parasitic" effects may be substantially reduced by subtracting a low irradiance background Z-scan from the high irradiance scan. A simple computer simulation of this process assuming that the surface imperfections do not disturb the circular symmetry of the beam or cause any beam steering, indicated that background subtraction indeed recovers the original ΔT_{p-v} arising from the nonlinear refraction effect even for quite large surface disturbances $\Delta\phi_s$ of up to π .

Returning to the Z-scan of Fig.7, we obtain $n_2 \simeq (0.8 \pm 0.15) \times 10^{-13}$ esu for BaF_2 at 532 nm, which is in close agreement with the reported values of 0.7×10^{-13} esu [21] and 1.0×10^{-13} esu [3] as measured at $1.06 \mu\text{m}$ using more complex techniques of nearly degenerate-three-wave-mixing and time-resolved-nonlinear-interferometry, respectively. Similarly for MgF_2 , we measure $n_2 \simeq 0.25 \times 10^{-13}$ esu at 532 nm as compared to the reported value of 0.32×10^{-13} esu at $1.06 \mu\text{m}$ for this material as given in [21]. Dispersion in n_2 for these materials between 1 and $0.5 \mu\text{m}$ is expected to be minimal. It should be noted that the n_2 values extracted from the Z-scans are absolute rather than relative measurements. If the beam parameters are not accurately known, however, it should be possible to calibrate the system by using a standard nonlinear material such as CS_2 .

5. CONCLUSION

We have demonstrated a simple single beam technique that is sensitive to less than $\lambda/300$ nonlinearly induced phase distortion. Using the Z-scan data the magnitude and sign of the nonlinear refraction can be simply determined. We have derived relations that allow the refractive index to be obtained directly from

the Z-scan data without resorting to computer fits. We have applied this technique to several materials displaying a variety of nonlinearities on different time scales. It is expected that this method will be a valuable tool for experimenters searching for highly nonlinear materials.

6. ACKNOWLEDGEMENT

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