High-sensitivity, single-beam n_2 measurements

M. Sheik-bahae, A. A. Said, and E. W. Van Stryland

Center for Research in Electro-Optics and Lasers, University of Central Florida, Orlando, Florida 32816

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We present a simple yet highly sensitive single-beam experimental technique for the determination of both the sign and magnitude of n_2. The sample is moved along the z direction of a focused Gaussian beam while the repetitively pulsed laser energy is held fixed. The resultant plot of transmittance through an aperture in the far field yields a dispersion-shaped curve from which n_2 is easily calculated. A transmittance change of 1% corresponds to a phase distortion of $\simeq\lambda/250$. We demonstrate this method on several materials using both CO_2 and Nd:YAG laser pulses.

Numerous techniques are known for the measurement of nonlinear refraction in materials. Nonlinear interferometry,[1,2] degenerate four-wave mixing,[3] nearly degenerate three-wave mixing,[4] ellipse rotation,[5] and beam-distortion measurements[6,7] are among the techniques frequently reported. The first three methods, interferometry and wave mixing, are potentially sensitive techniques but require a complex experimental apparatus. Beam-distortion measurements, on the other hand, require precise beam scans followed by detailed wave-propagation analysis. Based on the principles of spatial beam distortion, however, we present a single-beam technique for measuring the sign and magnitude of refractive nonlinearities that offers simplicity as well as high sensitivity. The technique is based on the transformation of phase distortion to amplitude distortion during beam propagation. We demonstrate this technique, which we refer to as a Z scan, on several materials in the IR and the visible, with nanosecond and picosecond pulses, for thermal and electronic Kerr nonlinearities. The demonstrated sensitivity to nonlinearly induced phase changes is better than $\lambda/100$.

The Z-scan experimental apparatus is shown in Fig. 1. Using a Gaussian laser beam in a tight-focus limiting geometry, we measure the transmittance of a nonlinear medium through a finite aperture placed in the far field as a function of the sample position (z) measured with respect to the focal plane. The following example qualitatively explains how such a trace (Z scan) is related to the nonlinear refraction of the sample. We place a thin material (i.e., with a thickness much less than the beam depth of focus) having $n_2 < 0$ well in front of the focus ($-z$ in Fig. 1). As the sample is moved toward the focus the increased irradiance leads to a negative lensing effect that tends to collimate the beam, thus increasing the aperture transmittance. With the sample on the $+z$ side of the focus, the negative lensing effect tends to augment diffraction, and the aperture transmittance is reduced. The approximate null at $z = 0$ is analogous to placing a thin lens at the focus that results in a minimal far-field pattern change. For still larger $+z$ the irradiance is reduced and the transmittance returns to the original linear value. We normalize this value to unity. A positive nonlinearity results in the opposite effect, i.e., lowered transmittance for the sample at negative z and enhanced transmittance at positive z. Induced beam broadening and narrowing of this type have been previously observed and explained for the case of band filling and plasma nonlinearities[8] and in the presence of nonlinear absorption in semiconductors.[9]

Not only is the sign of n_2 apparent from a Z scan, but the magnitude of n_2 can also be easily calculated using a simple analysis for a thin medium. Considering the geometry given in Fig. 1, we formulate and discuss a simple method of analyzing the Z scan. For a fast cubic nonlinearity the index of refraction is expressed in terms of nonlinear indices n_2 (esu) through

$$n = n_0 + \frac{n_2}{2}|E|^2 = n_0 + \Delta n, \tag{1}$$

where n_0 is the linear index of refraction and E is the electric field. Assuming a Gaussian beam traveling in the $+z$ direction, we can write the magnitude of E as

$$|E(r, z, t)| = |E_0(t)|\frac{w_0}{w(z)}\exp\left[-\frac{r^2}{w^2(z)}\right], \tag{2}$$

where $w^2(z) = w_0^2(1 + z^2/z_0^2)$ is the beam radius 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 air. E_0 denotes the radiation electric field at the focus and contains the temporal envelope of the laser pulse.

If the sample length is small enough such that changes in the beam diameter within the sample due to either diffraction or nonlinear refraction can be

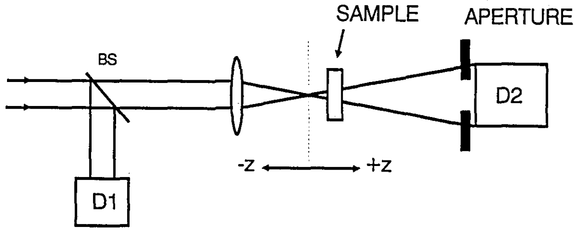

Fig. 1. Simple Z-scan experimental apparatus in which the transmittance ratio D_2/D_1 is recorded as a function of the sample position z. BS, Beam splitter.

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neglected, the medium is regarded as thin. Such an assumption simplifies the problem considerably, and the amplitude and nonlinear phase change $\Delta\phi$ of the electric field within the sample are now governed by

$$\frac{\mathrm{d}\Delta\phi}{\mathrm{d}z} = 2\pi/\lambda\ \Delta n \text{ and } \frac{\mathrm{d}|E|}{\mathrm{d}z} = -\alpha/2\ |E|, \quad (3)$$

where α is the linear absorption coefficient. Equations (3) 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:

$$\Delta\phi(r, z, t) = \frac{\Delta\Phi_0}{1 + z^2/z_0^2}\exp\left[-\frac{2r^2}{w^2(z)}\right], \quad (4a)$$

with

$$\Delta\Phi_0(t) = \frac{2\pi}{\lambda}\Delta n_0(t)\frac{1 - e^{-\alpha L}}{\alpha}, \quad (4b)$$

where L is the sample length and $\Delta n_0(t)$ is the instantaneous on-axis index change at the focus ($z = 0$). The electric field E' at the exit surface of the sample z_1 now contains the nonlinear phase distortion,

$$E'(r, z_1, t) = E(r, z_1, t)\exp(-\alpha L/2)\exp[i\Delta\phi(r, z_1, t)]. \quad (5)$$

By virtue of Huygens's principle one can obtain the far-field pattern of the beam at the aperture plane through a zeroth-order Hankel transformation of E'.[10] We use a numerically simpler Gaussian decomposition method given by Weaire *et al.*[11]

Having calculated the electric-field profile, E_a, at the aperture, we obtain the normalized instantaneous Z-scan power transmittance as

$$T(z, t) = \frac{\int_0^{r_a} |E_a(\Delta\Phi_0, r, z, t)|^2\, r\mathrm{d}r}{S\int_0^{\infty} |E_a(0, r, z, t)|^2\, r\mathrm{d}r}, \quad (6)$$

where r_a is the aperture radius and S is the aperture transmittance in the linear regime. The laser temporal pulse shape can be taken into account by simply performing a separate time integration on both the upper and lower terms in Eq. (6). This gives the Z-scan fluence transmittance $T(z)$. We first discuss the general features of the Z scan using a constant input field such that $T(z, t) = T(z)$.

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 is, however, an important parameter in that a larger aperture reduces the variations in $T(z)$, i.e., the sensitivity. This reduction is more prominent in the peak, where beam narrowing occurs, and results in a peak transmittance that cannot exceed $(1 - S)$. The effect vanishes for a large aperture or no aperture, where $S = 1$, and $T(z) = 1$ for all z and $\Delta\Phi_0$ (assuming no nonlinear absorption). For small $|\Delta\Phi|$, the peak and valley occur at the same distance with respect to the focus, and for a cubic nonlinearity their separation is found to be $\simeq 1.7z_0$. This distance may be used to determine the order of the nonlinearity.

We can define an easily measurable quantity ΔT_{p-v} as the difference between the normalized peak (maximum) and valley (minimum) transmittances, $T_p - T_v$. The variation of this quantity as a function of $\Delta\Phi_0$ as calculated for various aperture sizes is found to be almost linearly dependent on $\Delta\Phi_0$. Within ±3% accuracy the following relationship holds:

$$\Delta T_{p-v} \simeq p|\Delta\Phi_0| \text{ for } |\Delta\Phi_0| \le \pi, \quad (7a)$$

with $p = 0.405(1 - S)^{0.25}$. Particularly, for on-axis transmission ($S \simeq 0$) we find that

$$\Delta T_{p-v} \simeq 0.405|\Delta\Phi_0| \text{ for } |\Delta\Phi_0| \le \pi. \quad (7b)$$

The linear nature of relations (7) makes it convenient to account for the temporal and transient effects in Eq. (6) by simply averaging the instantaneous phase distortion $\Delta\Phi_0(t)$ over the laser pulse shape. An average phase distortion $\Delta\Phi_0$ can be obtained as the product of the peak phase shift $\Delta\Phi_0(0)$ and an averaging factor that is a constant of the pulse shape for a given type of nonlinearity. For example, for a Gaussian pulse shape and a fast cubic nonlinearity, this factor is $1/\sqrt{2}$. For a cumulative nonlinearity having a decay time much longer than the pulse width (e.g., thermal) a fluence averaging factor of 0.5 is to be used regardless of the shape of the pulse. Relations (7) can thus be used to calculate the nonlinear index n_2 to within ±3%. This equation also reveals the highly sensitive nature of the Z-scan technique. For example, if the experimental apparatus is capable of resolving transmittance changes (ΔT_{p-v}) of $\simeq$1%, phase changes corresponding to $\lambda/250$ wave-front distortion are detectable.

Figure 2 shows a Z scan of a 1-mm-thick CS_2 cell using 300-nsec pulses from a single-longitudinal-mode

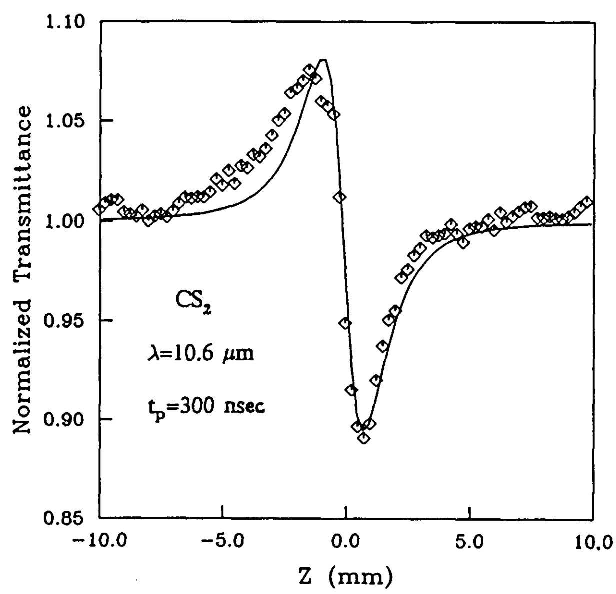

Fig. 2. Measured Z scan of a 1-mm-thick CS_2 cell using 300-nsec pulses at $\lambda = 10.6\ \mu m$ indicating thermal self-defocusing. The solid curve is the calculated result with $\Delta\Phi_0 = -0.6$.

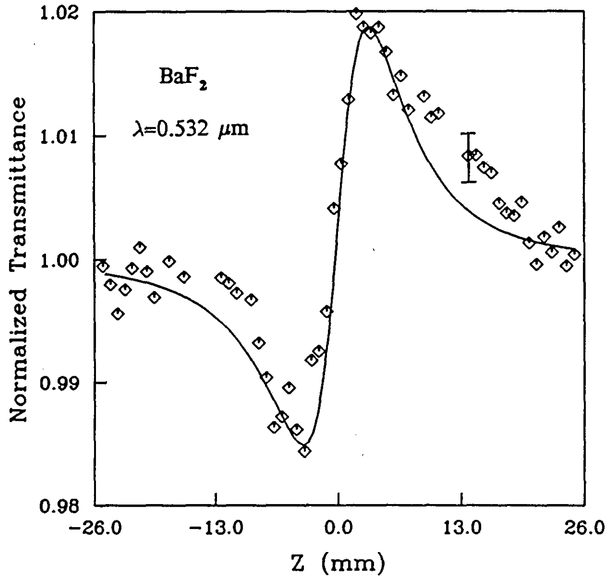

Fig. 3. Measured Z scan of a 2.5-mm-thick BaF_2 sample using 27-psec (FWHM) pulses at $\lambda = 0.532\ \mu m$ indicating the self-focusing due to the electronic Kerr effect. The solid curve is the theoretical fit with $\Delta\Phi_0 = 0.085$ corresponding to $\simeq\lambda/75$ phase distortion.

TEA CO_2 laser having an energy of 0.85 mJ. The peak-to-valley configuration of this Z scan is indicative of a negative (self-defocusing) nonlinearity. The solid curve in Fig. 2 is the calculated result using $\Delta\Phi_0 = -0.6$, which gives an index change of $\Delta n_0 = -1 \times 10^{-3}$. This is attributed to a thermal nonlinearity resulting from linear absorption of CS_2 ($\alpha \simeq 0.22\ cm^{-1}$) at 10.6 μm. The rise time 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. For CS_2 with $v_s \simeq 1.5 \times 10^5$ cm/sec and $w_0 \simeq 60\ \mu m$, we obtain a rise time of $\simeq$40 nsec, which is almost an order of magnitude smaller than the transversely excited atmosphere laser pulse width. Furthermore, the relaxation of the thermal lens, governed by thermal diffusion, is of the order of 100 msec.[12] Therefore we regard the nonuniform heating caused by the 300-nsec pulses as quasi-steady state, in which case the average on-axis nonlinear index change at focus can be determined in terms of the thermo-optic coefficient, dn/dT, as

$$\Delta n_0 \simeq \frac{dn}{dT}\frac{0.5F_0\alpha}{\rho C_v}, \qquad (8)$$

where F_0 is the fluence, ρ is the density, C_v is the specific heat, and 0.5 denotes the fluence averaging factor. With the known value of $\rho C_v \simeq 1.3$ J/K cm^3 for CS_2, we calculate that $dn/dT \simeq -(8.3 \pm 1.0) \times 10^{-4}$ $°C^{-1}$, which is in good agreement with the reported value of $-8 \times 10^{-4}\ °C^{-1}$.[13]

Using 27-psec (FWHM), 2.0-μJ pulses from a frequency-doubled Nd:YAG laser focused to a spot size w_0 of 18 μm, we performed a Z scan on a 2.5-mm-thick BaF_2 crystal. The result (Fig. 3) indicates a positive (self-focusing) nonlinearity. The theoretical fit assuming Gaussian-shaped pulses was obtained for $\Delta\Phi_0 = 0.085$, from which an n_2 value of $\simeq(0.8 \pm 0.15) \times 10^{-13}$ esu is calculated. This value is in agreement with the reported values of $\simeq 0.7 \times 10^{-13}$ and $\simeq 1.0 \times 10^{-13}$ esu as measured using nearly degenerate three-wave mixing[4] and time-resolved interferometry,[2] respectively. BaF_2 has a particularly small value of n_2. In addition, the laser input energy was purposely lowered to 2.0 μJ to illustrate the sensitivity of this technique to small induced phase changes. The peak wave-front distortion shown in Fig. 3 corresponds to λ/75.

The simplicity and sensitivity of the technique described here make it attractive as a screening test to give the sign, magnitude, and order of the nonlinear response of new nonlinear-optical materials.

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