Sensitive Measurement of Optical Nonlinearities Using a Single Beam

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Abstract—**We report a sensitive single-beam technique for measuring both the nonlinear refractive index and nonlinear absorption coefficient for a wide variety of materials. We describe the experimental details and present a comprehensive theoretical analysis including cases where nonlinear refraction is accompanied by nonlinear absorption. In these experiments, 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 nonlinear refraction are 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. In cases where nonlinear refraction is accompanied by nonlinear absorption, it is possible to separately evaluate the nonlinear refraction as well as the nonlinear absorption by performing a second Z scan with the aperture removed. We demonstrate this method for ZnSe at 532 nm where two-photon absorption is present and n_2 is negative.**

I. Introduction

RECENTLY we reported a single-beam method for measuring the sign and magnitude of n_2 that has a sensitivity comparable to interferometric methods [1]. Here, we describe this method in detail and demonstrate how it can be applied and analyzed for a variety of materials. We also extend this method to the measurement of nonlinear refraction in the presence of nonlinear absorption. Thus, this method allows a direct measurement of the nonlinear absorption coefficient. In addition, we 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], and beam distortion measurements [7], [8]. The first three methods, namely, nonlinear interferometry and wave mixing, are potentially sensitive techniques, but all require relatively complex experimental apparatus. Beam distortion measurements, on the other hand, are relatively insensitive and require detailed wave propagation analysis. The technique reported here is based on the principles of spatial beam distortion, but offers simplicity as well as very high sensitivity.

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We will describe this simple technique, referred to as a "Z-scan," 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 thermooptical and reorientational Kerr effects were identified using nanosecond and picosecond pulses, respectively. Furthermore, in Section V, we will consider the case of samples having a significant absorptive nonlinearity as well as a refractive one. This occurs in, for example, two-photon absorbing semiconductors. It will be shown that both effects can easily be separated and measured in the Z-scan scheme. We also show how effects of linear sample inhomogeneities (e.g., bulk index variations) can be effectively removed from the experimental data.

II. 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

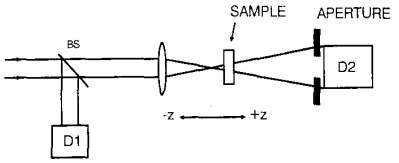

Fig. 1. The Z-scan experimental apparatus in which the ratio $D2/D1$ is recorded as a function of the sample position z.

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 by chemicals in solvents [11].

A prefocal transmittance maximum (peak) followed by a postfocal 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 can be extracted from such "open" aperture experiments. We will show in Section V how the data from the two Z-scans, with and without the aperture, can be used to separately determine both the nonlinear absorption and the nonlinear refraction. We will demonstrate this data analysis on semiconductors where two-photon absorption and self-refraction are simultaneously present.

III. 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, nonlinearities of any order 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 indexes n_2(esu) or $\gamma(\mathrm{m}^2/\mathrm{W})$ through

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

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 $n_2(\mathrm{esu}) = (cn_0/40\pi)\gamma(m^2/W)$ where $c(\mathrm{m/s})$ is the speed of light in vacuum.) Assuming a TEM_{00} Gaussian beam of beam waist radius w_0 traveling in the $+z$ direction, we can write E as

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

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" [14]. For linear diffraction, this implies that $L \ll z_0$, while for nonlinear refraction, $L \ll z_0/\Delta\phi(0)$. In most experiments using the Z-scan technique, we find that the second criterion is automatically met since $\Delta\phi$ is small. Additionally, we have found that the first criterion for linear diffraction is more restrictive than it need be, and it is sufficient to replace it with $L < z_0$. We have determined this empirically by measuring n_2 in the same material using various z_0's and the same analysis and have obtained the same value for n_2. 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:

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

and

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

where z' is the propagation depth in the sample and $\alpha(I)$, in general, includes linear and nonlinear absorption terms. Note that z' should not be confused with the sample position z. In the case of a cubic nonlinearity and negligible nonlinear absorption, (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,

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

with

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

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

$$\Delta\Phi_0(t) = k\Delta n_0(t) L_{\text{eff}} \quad (6)$$

where $L_{\text{eff}} = (1 - e^{-\alpha L})/\alpha$, with L the sample length and α the linear absorption coefficient. Here, $\Delta n_0 = \gamma I_0(t)$ with $I_0(t)$ being the on-axis irradiance at focus (i.e., $z = 0$). We ignore Fresnel reflection losses such that, for example, $I_0(t)$ is the irradiance within the sample.

The complex electric field exiting the sample E_e now contains the nonlinear phase distortion

$$E_e(r, z, t) = E(z, r, t)\, e^{-\alpha L/2}\, e^{i\Delta\phi(z,r,t)}. \quad (7)$$

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_e [15]. 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.* [14], 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 (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

$$E_a(r, t) = E(z, r = 0, t) e^{-\alpha L/2} \sum_{m=0}^{\infty} \frac{[i\Delta\phi_0(z, t)]^m}{m!} \frac{w_{m0}}{w_m} \cdot \exp\left(-\frac{r^2}{w_m^2} - \frac{ikr^2}{2R_m} + i\theta_m\right). \quad (9)$$

Defining d as the propagation distance in free space from the sample to the aperture plane and $g = 1 + d/R(z)$, the remaining parameters in (9) are expressed as

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

$$d_m = \frac{kw_{m0}^2}{2}$$

$$w_m^2 = w_{m0}^2\left[g^2 + \frac{d^2}{d_m^2}\right]$$

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

and

$$\theta_m = \tan^{-1}\left[\frac{d/d_m}{g}\right].$$

The expression given by (9) is a general case of that derived by Weaire *et al.* [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 (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

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

where ϵ_0 is the permittivity of vacuum. 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 $S = 1 - \exp(-2r_a^2/w_a^2)$ is the aperture linear transmittance, with w_a denoting the beam radius at the aperture in the linear regime.

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 $\Delta\Phi_0 = \pm 0.25$ 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 ($d >> z_0$) 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

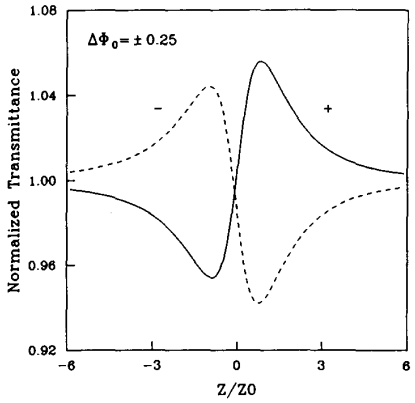

Fig. 2. Calculated Z-scan transmittance curves for a cubic nonlinearity with either polarity and a small aperture ($S = 0.01$).

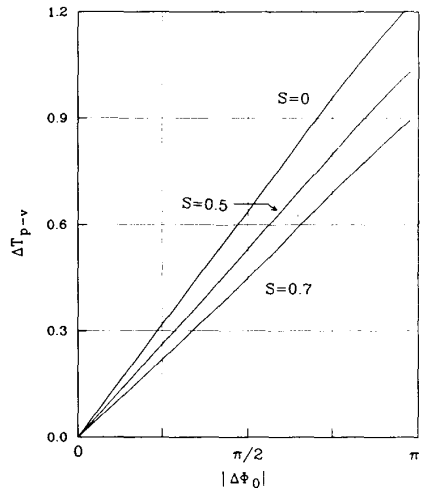

Fig. 3. Calculated ΔT_{p-v} as a function of the phase shift at the focus ($\Delta\Phi_0$). The sensitivity, as indicated by the slope of the curves, decreases slowly for larger aperture sizes ($S > 0$).

at the same distance with respect to focus, and for a cubic nonlinearity, this distance is found to be $\simeq 0.86\, z_0$ as shown in the Appendix. With larger phase distortions ($|\Delta\Phi_0| > 1$), numerical evaluation of (9)–(11) shows that this symmetry no longer holds and peak and valley both move toward $\pm z$ for the corresponding sign of nonlinearity ($\pm\Delta\Phi_0$) such that their separation remains nearly constant, given by

$$\Delta Z_{p-v} \simeq 1.7\, z_0. \tag{12}$$

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 almost linearly dependent on $|\Delta\Phi_0|$. As shown in the Appendix for small phase distortion and small aperture ($S \simeq 0$),

$$\Delta T_{p-v} \simeq 0.406\, |\Delta\Phi_0|. \tag{13a}$$

Numerical calculations show that this relation is accurate to within 0.5 percent for $|\Delta\Phi_0| \leq \pi$. As shown in Fig. 3, for larger apertures, the linear coefficient 0.406 decreases such that with $S = 0.5$, it becomes $\simeq 0.34$, and at $S = 0.7$, it reduces to $\simeq 0.29$. Based on a numerical fitting, the following relationship can be used to include such variations within a $\pm 2\%$ accuracy:

$$\Delta T_{p-v} \simeq 0.406(1 - S)^{0.25}\, |\Delta\Phi_0| \quad \text{for } |\Delta\Phi_0| \leq \pi. \tag{13b}$$

The implications of (13a) and (13b) 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 transmittance 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 results to include transient effects induced by pulsed radiation by using the time-averaged index change $\langle \Delta n_0(t) \rangle$ where

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

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

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

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 (e.g., thermal), the instantaneous index change is given by the following integral:

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

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

$$\langle \Delta n_0(t) \rangle = \frac{1}{2} AF \tag{17}$$

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

These equations were obtained based on a cubic nonlinearity (i.e., a $\chi^{(3)}$ effect). A similar analysis can be performed for higher order nonlinearities. Regardless of the order of the nonlinearity, the same qualitative features are to be expected from the Z-scan analysis. In particular, to quantify such features, we examined the effects of a $\chi^{(5)}$ nonlinearity which can be represented by a nonlinear index change given as $\Delta n = \eta I^2$. Nonlinearities encountered in semiconductors where the index of refraction is altered through charge carriers generated by two-photon absorption (i.e., a sequential $\chi^{(3)}:\chi^{(1)}$ effect) appear as such a fifth-order nonlinearity [20].

For a fifth-order effect, assuming a thin sample and using the GD approach, we find that the peak and valley are separated by $\simeq 1.2\, z_0$ as compared to $1.7\, z_0$ obtained for the third-order effect. Furthermore, the calculations also show that for a small aperture ($S \simeq 0$),

$$\Delta T_{p\text{-}v} \simeq 0.21\, |\Delta\Phi_0| \tag{18}$$

where, in this case, the phase distortion is given by

$$\Delta\Phi_0 = k\eta I^2 \left(\frac{1 - e^{-2\alpha L}}{2\alpha}\right). \tag{19}$$

Calculations also indicate that the aperture size dependence of (18) can be approximated by multiplying the right-hand term by $(1 - S)^{0.25}$, as was the case for a third-order nonlinearity.

As will be shown in Section V, we can also determine the nonlinear refraction in the presence of nonlinear absorption by separately measuring the nonlinear absorption in a Z-scan performed with the aperture removed. Within approximations elaborated in Section V, a simple division of the curves obtained from the two Z-scans will give the nonlinear refraction.

IV. 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 $\Delta T_{p\text{-}v}$ is needed). The defocusing effect shown in Fig. 4 is attributed to a thermal nonlinearity resulting from linear absorption of CS_2 ($\alpha \simeq 0.22\ \text{cm}^{-1}$ at 10.6 μm). The rise time of a thermal lens in a liquid is determined by the acoustic transit time $\tau \simeq w_0/v_s$ where v_s is the velocity of sound in the liquid [17]. For CS_2 with $v_s \simeq 1.5 \times 10^5$ cm/s and having $w_0 \simeq 60$ μm, we obtain a rise time of $\simeq 40$ 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 on 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 (17), the average on-axis nonlinear index change at focus can be determined in terms of the thermo-optic coefficient dn/dT as

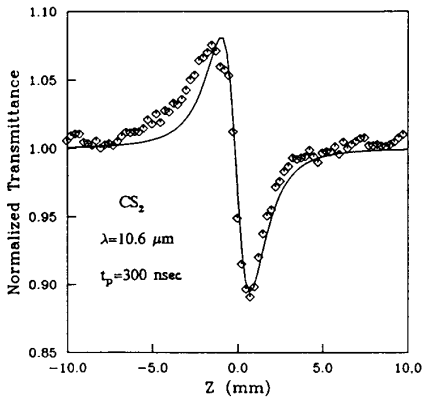

Fig. 4. Measured Z-scan of a 1 mm thick CS_2 cell using 300 ns pulses at $\lambda = 10.6$ μm indicating thermal self-defocusing. The solid line is the calculated result with $\langle \Delta\Phi_0 \rangle = -0.6$ and 60% aperture ($S = 0.6$).

$$\langle \Delta n_0 \rangle \simeq \frac{dn}{dT}\frac{F_0\alpha}{2\rho C_v} \tag{20}$$

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 $\rho C_v \simeq 1.3$ J/K · cm^3 for CS_2, we deduce $dn/dT \simeq -(8.3 \pm 1.0) \times 10^{-4}\ {}^0\text{C}^{-1}$, which is in good agreement with the reported value of $-8 \times 10^{-4}\ {}^0\text{C}^{-1}$ [16].

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, 1.06, and 0.53 μm to measure n_2 in CS_2. We obtain the same value of n_2, within errors, at all three wavelengths, $(1.5 \pm 0.6) \times 10^{-11}$ esu at 10.6 μm, $(1.3 \pm 0.3) \times 10^{-11}$ esu at 1.06 μm, and $(1.2 \pm 0.2) \times 10^{-11}$ esu at 0.53 μm. The external self-focusing arising from the Kerr effect in CS_2 is shown in Fig. 5 where a Z-scan of a 1 mm cell using 27 ps (FWHM) pulses focused to a beam waist w_0 of 25 μm from a frequency-doubled Nd:YAG laser is illustrated. Its valley–peak configuration indicates the positive sign of n_2. With $\Delta T_{p\text{-}v} = 0.24$, and using (13b) with a 40% 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 $n_2 \simeq (1.2 \pm 0.2) \times 10^{-11}$ esu. The main source of uncertainty in the value of n_2 is the absolute measurement of the irradiance. In this paper, all irradiance values quoted are values within the sample, i.e., including front surface reflection losses. A plot of $\Delta T_{p\text{-}v}$ versus peak laser irradiance as measured from various Z-scans on the same CS_2 cell is shown in Fig. 6. The linear behavior of this plot follows (13) as derived for a cubic nonlinearity.

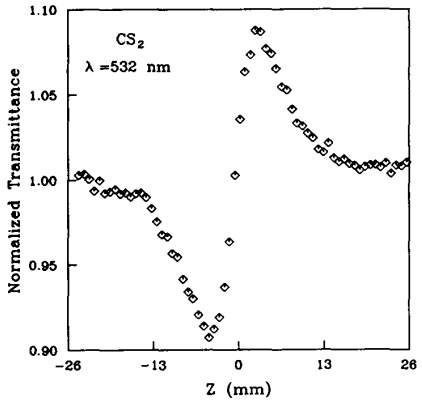

Fig. 5. Measured Z-scan of a 1 mm thick CS_2 cell using 27 ps pulses at λ = 532 nm. It depicts the self focusing effect due to the reorientational Kerr effect.

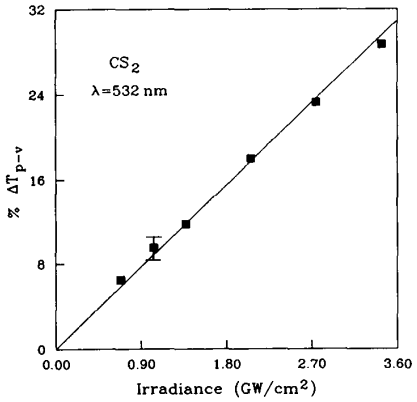

Fig. 6. ΔT_{p-v} in percent as a function of the peak irradiance from the Z-scan data of CS_2 at 532 nm, indicative of the reorientational Kerr effect.

Transparent dielectric window materials have relatively small nonlinear indexes. 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 $\lambda \simeq 1.06$ μm. Using the Z-scan technique, we examined some of these materials at 532 nm. For example, the result for a randomly oriented sample of BaF_2 (2.4 mm thick) is shown in Fig. 7, using the same beam parameters as for CS_2. This Z-scan was obtained with a 50% aperture and at a pulse energy of $\simeq 28$ μJ corresponding to a peak irradiance (I_0) of $\simeq 100$ GW/cm^2. A low irradiance (4 μJ) Z-scan of the same sample was shown in [1] to have a phase distortion resolution of better than $\lambda/300$. (The pulse energy for this Z-scan was misquoted as 2 μJ in [1].) Such a resolution is also shown in Fig. 7 by the arrows indicating the corresponding transmittance variation equal to the maximum scatter in the Z-scan data. For laser systems having better amplitude and pulsewidth stability, the sensitivity will be correspondingly improved.

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, after normalizing each scan. Fig. 8 shows Z-scan data before and after subtraction in a particularly poor 1 mm thick sample of ZnSe. 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, that is, $\Delta\phi_s$ of up to π.

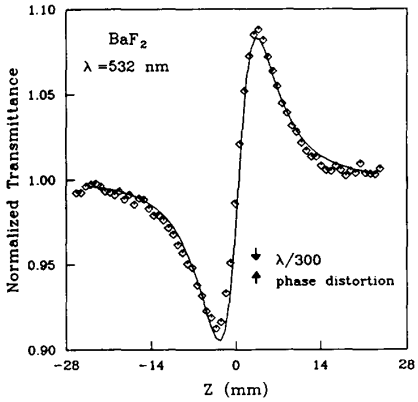

Fig. 7. Measured Z-scan of a 2.4 mm thick BaF_2 sample using 27 ps pulses at λ = 532 nm, indicating the self-focusing due to the electronic Kerr effect. The solid line is the calculated result with a peak $\Delta\Phi_0 = 0.73$. The separation of the arrows corresponds to an induced phase distortion of $\lambda/300$.

Returning to the Z-scan of Fig. 7, we obtain $n_2 \simeq (0.9 \pm 0.15) \times 10^{-3}$ esu for BaF_2 at 532 nm, which is in close agreement with our low irradiance measurement of $\simeq (0.8 \pm 0.15) \times 10^{-13}$ esu as reported in [1]. This compares well with other reported values of 0.7×10^{-13} esu [21] and 1.0×10^{-13} esu [3] as measured at 1.06 μm 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 μm for this material as given in [21]. Since the transparency region of these materials extends from mid-IR to UV, the dispersion in n_2 between 1 and 0.5 μm is expected to be negligible. 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.

V. Effects of Nonlinear Absorption

We now describe a method by which the Z-scan technique can be used to determine both the nonlinear refractive index and the nonlinear absorption coefficient for materials that show such nonlinearities simultaneously. Large refractive nonlinearities in materials are commonly associated with a resonant transition which may be of single or multiphoton nature. The nonlinear absorption in such

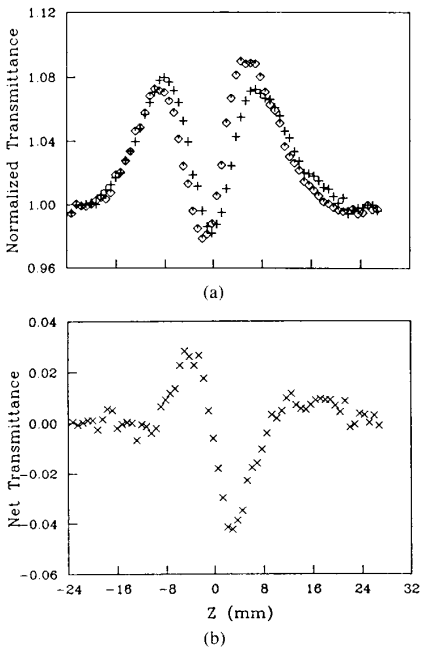

Fig. 8. (a) Measured Z-scans of a 1 mm thick ZnSe sample with poor surface quality for low irradiance (diamonds) showing the background and high irradiance (+). (b) Net transmittance change versus z after the background subtraction of the data in (a).

materials arising from either direct multiphoton absorption, saturation of the single photon absorption, or dynamic free-carrier absorption have strong effects on the measurements of nonlinear refraction using the Z-scan technique. Clearly, even with nonlinear absorption, a Z-scan with a fully open aperture ($S = 1$) is insensitive to nonlinear refraction (thin sample approximation). Such Z-scan traces with no aperture are expected to be symmetric with respect to the focus ($z = 0$) where they have a minimum transmittance (e.g., multiphoton absorption) or maximum transmittance (e.g., saturation of absorption). In fact, the coefficients of nonlinear absorption can be easily calculated from such transmittance curves.

Here, we analyze two-photon absorption (2PA), which we have studied in semiconductors with $E_g < 2\hbar\omega < 2E_g$ where E_g is the bandgap energy and ω is the optical frequency [22]. The third-order nonlinear susceptibility is now considered to be a complex quantity:

$$\chi^{(3)} = \chi_R^{(3)} + i\chi_I^{(3)} \tag{21}$$

where the imaginary part is related to the 2PA coefficient β through

$$\chi_I^{(3)} = \frac{n_0^2 \epsilon_0 c^2}{\omega} \beta \tag{22a}$$

and the real part is related to γ through

$$\chi_R^{(3)} = 2n_0^2 \epsilon_0 c \gamma. \tag{22b}$$

Here, we are concerned with the low excitation regimes where the free-carrier effects (refractive and absorptive) can be neglected. In view of this approximation, (3) and (4) will be reexamined after the following substitution:

$$\alpha(I) = \alpha + \beta I. \tag{23}$$

This yields the irradiance distribution and phase shift of the beam at the exit surface of the sample as

$$I_e(z, r, t) = \frac{I(z, r, t)\, e^{-\alpha L}}{1 + q(z, r, t)} \tag{24}$$

and

$$\Delta\phi(z, r, t) = \frac{k\gamma}{\beta} \ln [1 + q(z, r, t)] \tag{25}$$

where $q(z, r, t) = \beta I(z, r, t) L_{\text{eff}}$ (again, z is the sample position). Combining (24) and (25), we obtain the complex field at the exit surface of the sample to be [23]

$$E_e = E(z, r, t)\, e^{-\alpha L/2} (1 + q)^{(ik\gamma/\beta - 1/2)}. \tag{26}$$

Equation (26) reduces to (7) in the limit of no two-photon absorption. In general, a zeroth-order Hankel transform of (26) will give the field distribution at the aperture which can then be used in (10) and (11) to yield the transmittance. For $|q| < 1$, following a binomial series expansion in powers of q, (26) can be expressed as an infinite sum of Gaussian beams similar to the purely refractive case described in Section III as follows:

$$E_e = E(z, r, t)\, e^{-\alpha L/2} \sum_{m=0}^{\infty} \frac{q(z, r, t)^m}{m!} \cdot \left[\prod_{n=0} (ik\gamma/\beta - 1/2 - n + 1) \right] \tag{27}$$

where the Gaussian spatial profiles are implicit in $q(z, r, t)$ and $E(z, r, t)$. The complex field pattern at the aperture plane can be obtained in the same manner as before. The result can again be represented by (9) if we substitute the $(i\Delta\phi_0(z, t))^m/m!$ terms in the sum by

$$f_m = \frac{(i\Delta\phi_0(z, t))^m}{m!} \prod_{n=0}^{m} \left(1 + i(2n - 1)\frac{\beta}{2k\gamma}\right) \tag{28}$$

with $f_0 = 1$. Note that the coupling factor $\beta/2k\gamma$ is the ratio of the imaginary to real parts of the third-order nonlinear susceptibility $\chi^{(3)}$.

The Z-scan transmittance variations can be calculated following the same procedure as described previously. As is evident from (28), the absorptive and refractive contributions to the far-field beam profile and hence to the Z-scan transmittance are coupled. When the aperture is removed, however, the Z-scan transmittance is insensitive to beam distortion and is only a function of the nonlinear absorption. The total transmitted fluence in that case ($S = 1$) can be obtained by spatially integrating (24) without having to include the free-space propagation process. Integrating (24) at z over r, we obtain the transmitted power $P(z, t)$ as follows:

$$P(z, t) = P_i(t)\, e^{-\alpha L} \frac{\ln [1 + q_0(z, t)]}{q_0(z, t)} \tag{29}$$

where $q_0(z, t) = \beta I_0(t) L_{\text{eff}}/(1 + z^2/z_0^2)$ and $P_i(t)$ was

defined in (11). For a temporally Gaussian pulse, (29) can be time integrated to give the normalized energy transmittance

$$T(z, S = 1) = \frac{1}{\sqrt{\pi} q_0(z, 0)} \cdot \int_{-\infty}^{\infty} \ln\left[1 + q_0(z, 0)\, e^{-\tau^2}\right] d\tau. \quad (30)$$

For $|q_0| < 1$, this transmittance can be expressed in terms of the peak irradiance in a summation form more suitable for numerical evaluation:

$$T(z, S = 1) = \sum_{m=0}^{\infty} \frac{\left[-q_0(z, 0)\right]^m}{(m + 1)^{3/2}}. \quad (31)$$

Thus, once an open aperture ($S = 1$) Z-scan is performed, the nonlinear absorption coefficient β can be unambiguously deduced. With β known, the Z-scan with aperture in place ($S < 1$) can be used to extract the remaining unknown, namely, the coefficient γ.

An experimental example of this procedure is shown in Fig. 9 where a 2.7 mm thick ZnSe sample is examined using 27 ps (FWHM) pulses at 532 nm. ZnSe with a bandgap energy of 2.67 eV is a two-photon absorber at this wavelength. With a linear index of 2.7, the diffraction length inside the sample ($n_0 z_0$) was approximately four times the sample thickness. This allows us to safely apply the thin sample analysis developed in this paper. Fig. 9(a) depicts the open aperture data at a peak irradiance I_0 of 0.21 GW/cm^2. Also plotted is the theoretical result using (28) in (9) with $\beta = 5.8$ cm/GW. This is in excellent agreement with the previously measured value of 5.5 cm/GW [22]. Under the same conditions, the Z-scan with a 40% aperture, as shown in Fig. 9(b), exhibits a self-defocusing effect. These data have had a low irradiance background Z-scan subtracted to reduce the effects of linear sample inhomogeneities. Note the significant difference between this Z-scan and that of a purely refractive case. Here, the nonlinear absorption (2PA) has greatly suppressed the peak and enhanced the valley of the transmittance. The theoretical fit in Fig. 9(b) is obtained by setting $\beta = 5.8$ cm/GW and adjusting γ to be 6.8×10^{-14} cm^2/W ($n_2 = 4.4 \times 10^{-11}$ esu) with an uncertainty of $\pm 25\%$ arising predominantly from the irradiance calibration.

An irradiance-dependent Z-scan study of the ZnSe indicates that for an irradiance $I_0 < 0.5$ GW/cm^2, the nonlinear refraction is dominated by a third-order effect. This is depicted in Fig. 10 where the measured nonlinear index change Δn_0 varies linearly with the irradiance. At higher irradiance levels, however, the nonlinear refraction caused by 2PA generated charge carriers, an effective fifth-order nonlinearity, becomes important. This is indicated in Fig. 10 by the small deviation of Δn_0 at $I_0 = 0.57$ GW/cm^2 from the line representing the cubic nonlinearity. An earlier investigation of ZnSe using picosecond time-resolved degenerate four-wave mixing (DFWM) at 532 nm had indicated that a fast $\chi^{(3)}$ effect followed by a slowly decaying $\chi^{(5)}_{\text{eff}}$ resulting from two-photon generated charge carriers was responsible for the DFWM signal [24]. Z-scan experiments reported here verify those results, and in addition, can accurately determine the sign and magnitude of these nonlinearities.

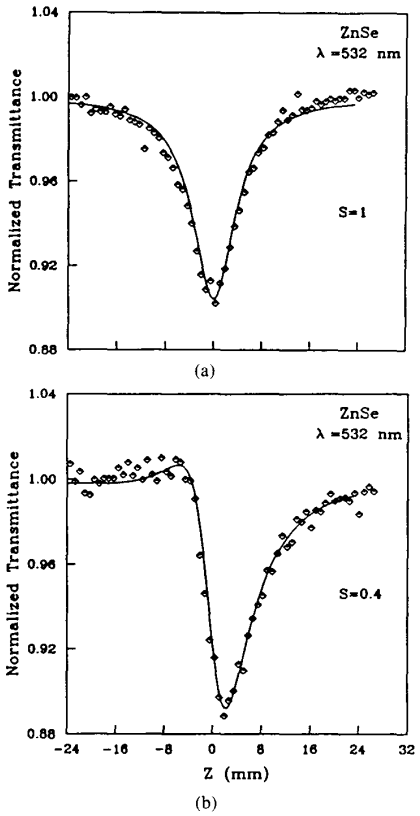

Fig. 9. Normalized Z-scan transmittance of ZnSe measured using picosecond pulses at $\lambda = 532$ nm with $I_0 = 0.21$ GW/cm^2. The solid lines are the theoretical results. (a) No aperture ($S = 1$) data and fit using 5.8 cm/GW. (b) 40% aperture data fitted with $\beta = 5.8$ cm/GW and $\gamma = 6.8 \times 10^{-5}$ cm^2/GW.

As was done for the case of a purely refractive effect, it is desirable to be able to estimate γ and β without having to perform a detailed fitting of the experimental data. A thorough numerical evaluation of the theoretical results derived in this section indicated that within less than 10% uncertainty, such a procedure is possible provided that $q_0(0, 0) \leq 1$ and $\beta/2k\gamma \leq 1$. The first condition can be met by adjusting the irradiance. The second condition is an intrinsic property of the material implying that the $\text{Im}(\chi^{(3)})$ should not be larger than the $\text{Re}(\chi^{(3)})$. This is the case for the semiconductors studied as well as for a wide variety of other materials. The separation and evaluation process is simple: divide the closed aperture ($S < 1$) normalized Z-scan (with background subtracted) by the one with open aperture ($S = 1$). The result is a new Z-scan where ΔT_{p-v} agrees to within $\pm 10\%$ of that obtained from a purely refractive Z-scan. The result of this procedure for the Z-scans of Fig. 9 is illustrated in Fig. 11 where the division of the two Z-scans of both experiment and theory are compared to the calculated Z-scan with β

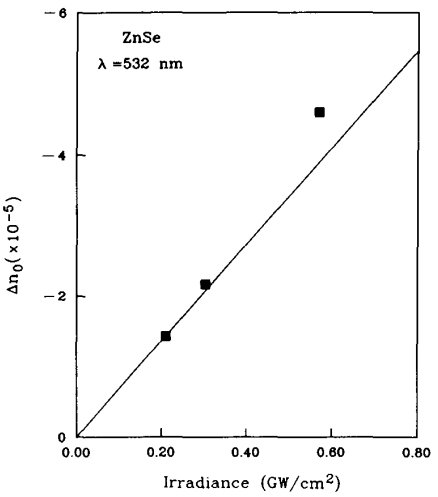

Fig. 10. The change of index in ZnSe versus the peak irradiance I_0 as measured from the Z-scan experiments. The line represents a cubic (n_2 type) nonlinearity. The deviation from the line is indicative of higher order refractive effects arising from two-photon generated charge carriers. The negative sign of the index change is apparent from the peak-valley configuration of Fig. 9(b).

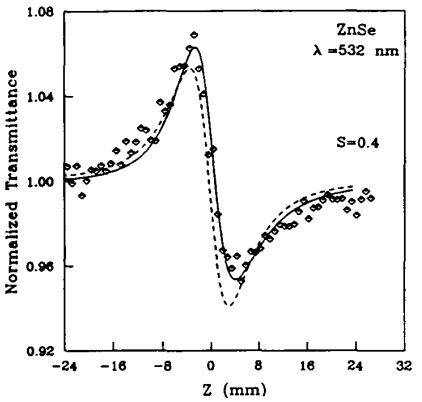

Fig. 11. The result of the division of the Z-scans of Fig. 9 (b)/(a): experimental (diamonds) and theoretical (solid line). The broken line shows the calculated result assuming $\beta = 0$. The ΔT_{p-v} of the latter agrees with that of the solid line fit to within 3%, making it possible to quickly estimate γ.

$= 0$. A simple measurement of ΔT_{p-v} and using (13) readily gives a value of $\gamma = 6.7 \times 10^{-14}$ cm^2/W, which is in excellent agreement with the value 6.8×10^{-14} cm^2/W obtained earlier.

VI. 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 of the nonlinear absorption and the magnitude and sign of the nonlinear refraction can be separately determined. We have derived simple 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.

Appendix

Here, we derive the on-axis Z-scan transmittance for a cubic nonlinearity and a small phase change. The on-axis electric field at the aperture plane can be obtained by letting $r = 0$ in (9). Furthermore, in the limit of small nonlinear phase change ($|\Delta\Phi_0| << 1$), only two terms in the sum in (9) need be retained. Following such simplifications, the normalized Z-scan transmittance can be written as

$$T(z, \Delta\Phi_0) = \frac{\left|E_a(z, r = 0, \Delta\phi_0)\right|^2}{\left|E_a(z, r = 0, \Delta\phi_0 = 0)\right|^2} = \frac{\left|(g + i\,d/d_0)^{-1} + i\Delta\phi_0(g + i\,d/d_1)^{-1}\right|^2}{\left|(g + id/d_0)^{-1}\right|^2}. \quad \text{(A1)}$$

The far-field condition $d >> z_0$ can be used to further simplify (A1) to give a geometry-independent normalized transmittance as

$$T(z, \Delta\Phi_0) \simeq 1 - \frac{4\Delta\Phi_0 x}{(x^2 + 9)(x^2 + 1)} \quad \text{(A2)}$$

where $x = z/z_0$.

The extrema (peak and valley) of the Z-scan transmittance can be calculated by solving the equation $dT(z, \Delta\Phi_0)/dz = 0$. Solutions to this equation yield

$$x_{p,v} = \pm\sqrt{\frac{\sqrt{52} - 5}{3}} \simeq \pm 0.858. \quad \text{(A3)}$$

Therefore, we can write the peak-valley separation as

$$\Delta Z_{p-v} \simeq 1.7\, z_0. \quad \text{(A4)}$$

Also, inserting the x values from (A3) into (A2), the peak-valley transmittance change is

$$\Delta T_{p-v} = \frac{8\left|x_{p,v}\right|}{(x_{p,v}^2 + 9)(x_{p,v}^2 + 1)}\Delta\Phi_0 = 0.406\,\Delta\Phi_0. \quad \text{(A5)}$$

Acknowledgment

We wish to thank A. Miller and M. J.Soileau for helpful discussions.

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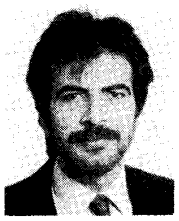

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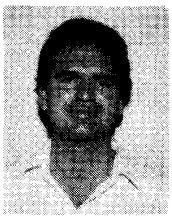

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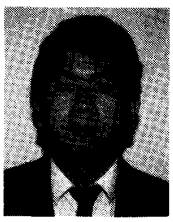

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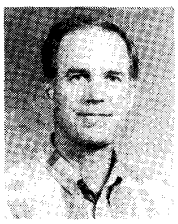
Dr. Hagan is currently President of the Florida Section of the Optical Society of America and is also a member of the Society of Photo-optical Instrumentation Engineers.



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