Eclipsing Z-scan measurement of $\lambda/10^4$ wave-front distortion

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We introduce a simple modification to the Z-scan technique that results in a sensitivity enhancement that permits measurement of nonlinearly induced wave-front distortion of $\simeq\lambda/10^4$. This sensitivity was achieved with 10-Hz repetition-rate pulsed laser sources. Sensitivity to nonlinear absorption is also enhanced by a factor of $\simeq 3$. This method permits characterization of nonlinear thin films without the need for waveguiding.

Since the introduction of the Z scan,[1] a sensitive single-beam technique for measuring nonlinear refraction (NLR), several variations have been introduced to enhance the technique. These include measurements in the presence of nonlinear absorption[2] (NLA), the two-color Z scan for the study of nondegenerate nonlinearities,[3,4] the time-resolved Z scan,[4,5] and measurements of the anisotropy of NLR.[6] Most of these experiments have been performed with low-repetition-rate ($\simeq$10-Hz) picosecond or nanosecond laser systems. Even with these low data-acquisition rates, the technique has demonstrated a sensitivity to wave-front distortion of $\lambda/300$ for a signal-to-noise ratio (S/N) of unity. It has been shown theoretically that a threefold enhancement of the Z scan's sensitivity to NLR can be achieved by the use of a lens between the sample and the aperture,[7] but this has yet to be realized experimentally. Recently it was demonstrated that the use of a top-hat beam profile in the Z scan results in an increase in sensitivity to NLR of $\simeq$2.5.[8] Here we introduce a simple variation of the Z-scan technique that provides greater than an order-of-magnitude enhancement of the S/N. This modification involves replacing the far-field aperture used in the standard Z scan with an obscuration disk that blocks most of the beam. The resulting pattern of light that passes around the edge of the disk, shown in the inset of Fig. 1, appears as a thin halo of light, reminiscent of a solar eclipse; hence this technique is named the eclipsing Z scan (EZ scan). This modification of the Z scan, accompanied by methods to compensate for fluctuations of the beam spatial profile, results in a sensitivity to induced wave-front distortion of $\simeq\lambda/10^4$ with a S/N of unity from a 10-Hz repetition-rate pulsed laser. Significantly higher sensitivities should be possible for more stable or higher-repetition-rate laser systems.

The EZ-scan experimental setup is shown in Fig. 1, where the aperture of the Z scan has been replaced by an opaque disk in the far field. As with the Z scan, a thin nonlinear sample is scanned along the Z axis of a focused Gaussian beam. In the case of a self-focusing nonlinearity ($n_2 > 0$, where $n = n_0 + n_2 I$), the sample will behave as a positive lens near the focus. Thus, for the sample positioned prior to focus, the far-field beam divergence is increased, and more light will pass by the disk in the far field. Note that this is exactly opposite the decreased transmittance of the aperture for a Z scan. With the sample positioned after the focal plane, the effect of the sample is to collimate the beam, and the disk blocks more of the light. Consequently, in the EZ scan a self-focusing medium results in an increase in transmittance (peak), followed by a decrease (valley) as the sample is scanned from in front of to behind the focus. For self-defocusing media, the positions of the valley and peak are reversed.

For a thin sample[9] this behavior can be modeled by the separated equations for irradiance, I, and induced phase shift, $\Delta\phi$: $\mathrm{d}I/\mathrm{d}z' = -\alpha I$ and $\mathrm{d}\Delta\phi/\mathrm{d}z' = k n_2 I(z')$, where α is the linear absorption coefficient, $k = 2\pi/\lambda$, and λ is the wavelength in vacuum. z' is the depth within the sample, as distinct from Z, the sample position with respect to the beam waist. In our modeling we assume the incident beam to be Gaussian. The integrated phase shift for a sample of length L follows the radial variation of the

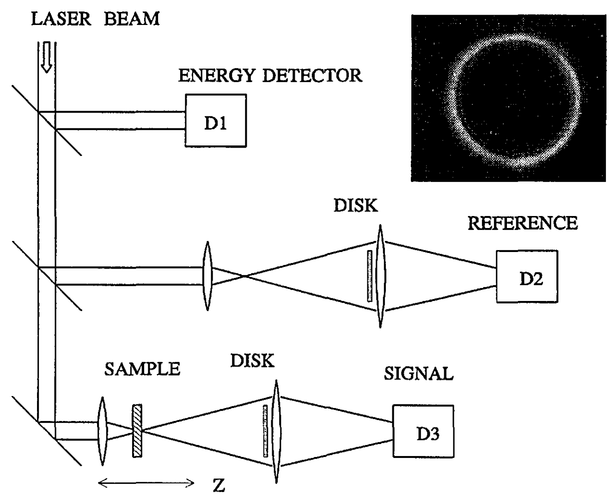

Fig. 1. Experimental arrangement for the EZ scan. D1 is the input energy monitor, and D3 measures the energy transmitted through the sample and past the disk. D2 monitors the energy transmitted through the reference arm, which is identical to the signal arm with no sample. The measured quantity is the ratio D3/D2. Inset: CCD image of a picosecond ND:YAG laser beam after it passes a 99% obscuration disk.

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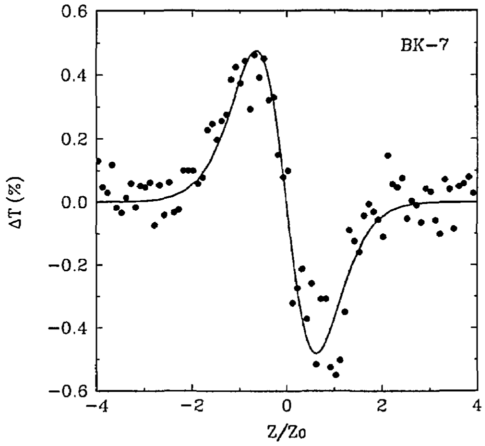

Fig. 2. EZ scan of a 1.5-mm-thick BK-7 sample performed with a frequency-doubled picosecond Nd:YAG laser. The solid curve is a fit to the data, indicating a peak wave-front distortion of $\lambda/2200$ with a S/N of approximately 5.

incident irradiance, $I \propto |E|^2$, for each sample position Z. Hence $\Delta\phi(r, Z, t) = \Delta\Phi_0(t)|E(r, Z, t)/E(0, 0, t)|^2$, where $\Delta\Phi_0(t) = \Delta\phi(0, 0, t) = kn_2 I(0, 0, t)L_{\text{eff}}$ is the on-axis phase shift at focus and $L_{\text{eff}} = [1 - \exp(-\alpha L)]/\alpha$. The electric field inside the medium at the exit surface is given by $E'(r, Z, t) = E(r, Z, t)\exp[-\alpha L/2 + i\Delta\phi(r, Z, t)]$, and the irradiance distribution at the plane of the disk (or aperture) can be found through a diffraction calculation or by the Gaussian decomposition method of Weaire *et al.*[9]

Figure 2 shows an EZ scan of a 1.5-mm-thick sample of BK-7 glass in which the normalized transmittance change, ΔT, is plotted as a function of Z. The laser source is a hybridly mode-locked and Q-switched frequency-doubled Nd:YAG laser emitting single 28-ps (FWHM) pulses at 532 nm with the input energy purposely reduced to less than 50 nJ to show the system noise. The beam was focused to a spot size of 14 μm (half-width at $1/e^2$ intensity), resulting in a peak irradiance of 0.52 GW/cm^2. Small beam-shape or beam-pointing fluctuations cause significant noise in the signal as measured by the ratio of energies detected by D3 and D1 (see Fig. 1). This led to the introduction of the reference arm shown in Fig. 1 (first proposed by Ma *et al.* for the Z scan[4]), which propagates a portion of the beam along an identical optical path except without a sample. By taking the ratio of energies D3 and D2, we can increase the S/N by a factor of 3–5. In our system, with the 100-shot average per data point shown in Fig. 2, the rms noise can be reduced to $\pm 0.1\%$ with this method. The solid curve fitted to the data in Fig. 2 gives $\Delta\Phi_0 \simeq 2\pi/2200$ for S/N $\simeq 5$. Thus with a S/N of unity we can resolve $\Delta\Phi_0 < 2\pi/10^4$, corresponding to a physical displacement of the wave front of $< \lambda/10^4$ or 0.05 nm. Using the same system with the reference arm, but performing a Z scan, we find that the sensitivity is limited to $\simeq\lambda/700$. The increased sensitivity of the EZ scan is due to the larger fractional change in irradiance in the wings of the beam that are detected in an EZ scan compared with that near the center, as detected in a Z scan. We conservatively define S/N $= \Delta T_{\text{pv}}/4\sigma$, where ΔT_{pv} is the difference between normalized peak and valley transmittances,[1] and σ is the standard deviation of the data in the absence of a nonlinearity.

For the Z scan,[1] ΔT_{pv} is almost linearly related to the light-induced phase shift at the focus, $\Delta\Phi_0$,

$$\Delta T_{\text{pv}} \simeq 0.406(1 - S)^{0.25}|\Delta\Phi_0|, \qquad (1)$$

where S is the aperture transmittance. This relationship holds to within $\pm 3\%$ for phase shifts $\Delta\Phi_0 < \pi$. With equal disk and aperture sizes, the corresponding absolute changes in transmitted power or energy are equal and opposite. However, the fractional transmittance changes may differ greatly for aperture and disk since much less light is transmitted with the disk for S near unity. In Fig. 3 we show the calculated ΔT_{pv} versus the disk radius and versus the aperture radius a for the EZ scan and the Z scan, respectively, for $\Delta\Phi_0 = 0.1$. The fraction of light blocked by the disk is simply S, the aperture transmittance in a Z scan, which is $S = 1 - \exp(-2a^2/w_a{}^2)$, where w_a is the beam radius at the disk plane in the linear regime.[1,2] We see from Fig. 3 that for large enhancement to be obtained, S must be within a few percent of unity. In practice this limits the maximum sensitivity enhancement as the energy reaching the detector becomes too small to detect. For our system $S = 0.99$, as used in the inset in Fig. 1, gave good enhancement while still giving sufficient energy for easy detection. Note that for $S = 0.5$, corresponding to $a = w_a\sqrt{2\ln 2}$, the sensitivities of the Z scan and EZ scan are identical, as expected.

For a large disk, $0.995 > S > 0.98$ (the useful range for this technique), and a small nonlinear phase shift $\Delta\Phi_0 \le 0.2$, we find a similar empirical linear relationship between ΔT_{pv} and $\Delta\Phi_0$ for the EZ scan:

$$\Delta T_{\text{pv}} \simeq 0.68(1 - S)^{-0.44}|\Delta\Phi_0|, \qquad (2)$$

which is accurate to within $\pm 3\%$. For the above range of S the spacing between the peak and valley,

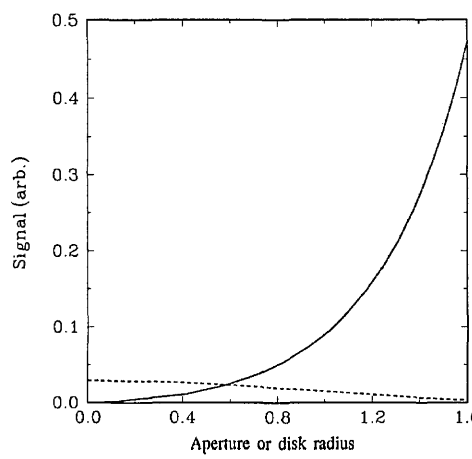

Fig. 3. Calculated ΔT_{pv} for an EZ scan (solid curve) and a Z scan (dashed curve) versus the normalized aperture or disk radius, respectively, for a peak on-axis phase shift of $|\Delta\Phi_0| = 0.1$.

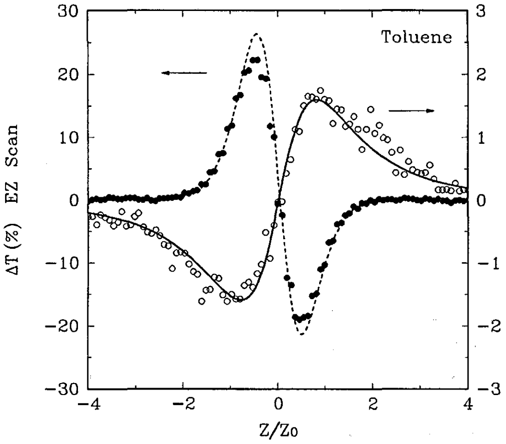

Fig. 4. Experimental comparison of an EZ scan (filled circles) and a Z scan (open circles) of toluene under identical conditions. Note that the Z-scan data are plotted on a 10× expanded scale for clarity. The laser source is a frequency-doubled nanosecond Nd:YAG laser. The solid and dashed curves are the result of calculations with a common $\Delta\Phi_0$ obtained from a best fit to the Z scan only.

ΔZ_{pv}, is empirically found to be given by $\Delta Z_{pv} = 0.9Z_0 - 1.0Z_0$ which grows to the Z-scan value of $\simeq 1.7Z_0$ as $S \rightarrow 0$. As described in Ref. 1, the linearity of relations (1) and (2) allows for simple analysis of pulsed laser experiments, as we may simply integrate over the laser pulse shape to get the normalized energy transmittance change. Thus, for pulsed sources, $\Delta\Phi_0$ in relation (2) should be replaced by its time-averaged value.[1,2]

Figure 4 shows an EZ scan and a Z scan performed under identical conditions on a 1-mm-thick cuvette filled with toluene. Note that the vertical scale for the Z scan has been expanded by a factor of 10 for clarity. Both scans were performed with the second harmonic of a single-longitudinal-mode Q-switched Nd:YAG laser operating in the TEM_{00} mode. The $\lambda = 0.532$ nm, 4.7-ns (FWHM) pulse was focused to a beam radius of 22 μm (half-width at $1/e^2$ intensity). In each case the incident energy was 62 μJ, corresponding to a peak irradiance at the beam waist of 1.68 GW/cm^2. The EZ scan used a disk of $S = 0.99$, while for the Z scan an aperture of $S = 0.40$ was used. The solid and dashed curves are fits performed with the thin sample approximation, in each case using the same $\Delta\Phi_0$ as determined from the Z scan. Experimentally, we observe a factor-of-13 increase in sensitivity for the EZ scan, compared with a predicted improvement of 15. This difference may be due to deviations from a perfect Gaussian beam profile or slight misalignment of the disk. Errors that are due to deviations from $S = 0.99$ are small, as the estimated error in $(1 - S)$ is $<\pm 5\%$, which from relation (2) results in an error for $\Delta\Phi_0$ of $\simeq\pm 2\%$.

As with the Z scan, one may also study samples exhibiting both NLR and NLA by performing successive EZ scans with and without the disk. Removing the disk gives an open-aperture Z scan that is sensitive only to nonlinear losses.[2] Interestingly, the presence of a far-field obscuration disk also results in an increase in sensitivity to NLA. For example, in the case of reverse saturable absorption or two-photon absorption the center portion of the beam is more strongly absorbed than the wings, thereby spatially broadening the beam as it leaves the sample. Propagation transforms this near-field broadening into far-field narrowing, causing more of the beam to be blocked by the disk and enhancing the fractional change in transmittance seen by the detector. For similar reasons, an obscuration disk also enhances the effect of saturable absorption.

In summary, we have demonstrated that the EZ scan, in combination with beam fluctuation compensation, provides a highly sensitive method for measuring small nonlinearly induced phase shifts, while retaining the ability to discriminate between NLR and NLA, and for determining the sign of each of these effects. The method is particularly relevant to the current problem of determining nonresonant nonlinearities in thin films without the need for waveguide coupling. For films of thickness $d \simeq \lambda$, a sensitivity to wave-front distortion of $\lambda/10^4$ corresponds to a sensitivity to index changes of $\Delta n \simeq 10^{-4}$.

As we observed from Fig. 4, the enhancement in sensitivity comes at the expense of a reduction in accuracy caused, we believe, be deviations from a Gaussian irradiance distribution. We therefore recommend use of this technique with a known reference to calibrate the system (for our system, without such calibration, the absolute accuracy was within 18%). The Z scan is still the method of choice unless the S/N is a problem.

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