Z-scan measurement technique for non-Gaussian beams and arbitrary sample thicknesses

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We demonstrate a new Z-scan measurement technique that permits the use of non-Gaussian beams and thick, as well as thin, samples. We expect that this technique will make possible the measurement of optical nonlinearities by the use of lasers that previously would have been unsuitable for this purpose, because of either inadequate beam quality or inadequate power. Another advantage of this technique is that it does not require detailed knowledge of the temporal characteristics of the laser pulse that is used. © 1995 Optical Society of America

The Z-scan measurement technique is a simple experimental procedure that gives information on the optical nonlinearities of materials. The technique as originally formulated[1,2] is performed by sending an axially symmetric Gaussian beam through a converging lens, then through a sample of material placed near the beam waist, and finally through an aperture placed in front of a detector in the far field. As the sample is moved to one side of the beam waist, the detected power increases to a peak; as the sample is moved to the other side of the waist, the detected power decreases to a valley. The difference in power from the peak to the valley has been shown to be proportional to the nonlinear index of refraction n_2. Consequently the Z-scan technique permits determination of n_2 for different materials.

In its original formulation, the Z-scan technique assumes that the input beam is Gaussian (with a beam-quality factor[3] of $M^2 = 1$). Often, however, the lasers found in laboratories do not produce Gaussian beams. The Nd:YAG laser, which is often used in Z-scan measurements, may have a beam-quality factor of $M^2 > 2$.[4] It is possible to modify a laser beam to make it a more Gaussian; for example, one group obtained $M^2 = 1.02$ by sending light from a doubled Nd:YAG laser through a spatial filter and an apodizer.[5] Such modifications are not always practical or convenient, however. An alternative to the Gaussian beam is the top-hat beam,[6] which can be obtained by use of a spatial filter, an expander, and an aperture. Much of the pulse energy may be thrown away in forming a top-hat beam, but, in partial compensation, the peak-to-valley response is increased by a factor of 2.5 compared with that of a Gaussian beam.[6]

The original formulation also assumes that the sample is much thinner than a Rayleigh range.[2] For a laser of moderate pulse power, this condition may limit the minimum value of n_2 that can be measured. This is the case because the peak-to-valley change in transmittance decreases as the sample thickness is made smaller. More recently, techniques have been developed for applying the Z-scan method to samples thicker than a Rayleigh range. One of these techniques is based on empirical observations,[7] and another is based on a (more accurate) Gaussian–Laguerre decomposition method.[5] The Z-scan measurement techniques described above also require knowledge of the temporal profile of the laser pulse for accurate calculation of n_2,[5] but in some cases this information may not be easy to obtain.

In this Letter, we present a new Z-scan measurement technique that permits the use of lasers that do not have ideal Gaussian beams. This technique permits the use of thick or thin samples and can be used to determine n_2 without detailed knowledge of the temporal profile of the laser pulse. To understand the new measurement procedure, we now derive a differential equation that accounts for the physical effects of interest. We begin by defining the electric field E in terms of the electric field amplitude A,

$$E(\mathbf{r}, T) = \tfrac{1}{2}A(\mathbf{r}, T)\exp[i\beta_0^{(j)}z - i\omega_0 T] + \text{c.c.}, \quad (1)$$

where T is the time in the laboratory frame of reference, $\beta_0^{(j)} = n_0^{(j)}k_0$ is the propagation constant, $k_0 = \omega_0/c$ is the vacuum propagation constant at the reference frequency ω_0, and $n_0^{(j)}$ is the linear index of refraction. The superscript (or subscript) j is used to identify each different material. We assume that linear absorption and two-photon absorption are negligible. We also assume that temporal dispersion has a negligible effect, which is usually a very good assumption for pulse lengths greater than 1 ps. For an intensity-dependent change in the refractive index of the jth sample of $\Delta n_j = n_2^{\prime(j)}|A|^2/2$, the differential equation that governs the propagation of the beam through the Z-scan test setup is then[8]

$$i\frac{\partial}{\partial z}A = -\frac{1}{2\beta_0^{(j)}}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)A - \frac{\omega_0 n_2^{\prime(j)}|A|^2}{2c}A, \quad (2)$$

where $A = A(x, y, t; z)$, with t the time referenced to the center of the pulse and z the propagation direction. The first term on the right-hand side of Eq. (2) accounts for diffraction in the x and y directions.

Using SI units and the conventions of Eq. (1), $|A(x, y, t; z)|^2 = 2I(x, y, t; z)/n_0^{(j)}\epsilon_0 c$,[9] where I is

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the intensity. We use this result to evaluate the quantity $N(t) = \iint_{-\infty}^{\infty} dx dy |A(x, y, t; z)|^2$ to obtain $N(t) = 2P(t)/n_0^{(j)} \epsilon_0 c$, where $P(t)$ in the instantaneous power. Note that N and P are functions of t but not of z because temporal dispersion and loss are assumed negligible. The coefficient $n_2^{(j)}$, defined by the relation $\Delta n_j = n_2^{(j)} I$, is related to the coefficient $n_2'^{(j)}$ by $n_2'^{(j)} = n_0^{(j)} \epsilon_0 c n_2^{(j)}$, from which it follows that $n_2'^{(j)} N = 2 n_2^{(j)} P$. We use this, along with the definition of the critical power, $P_{cj} = 2\pi/k_0^2 n_0^{(j)} n_2^{(j)}$,[10] and the definition of the normalized field amplitude, $u(x, y; z) = A(x, y, t; z)/\sqrt{N(t)}$, to rewrite the nonlinear term in Eq. (2) as $[2\pi P/\beta_0^{(j)} P_{cj}]|u|^2$. (Note that P_{cj} as defined can be negative.) We substitute this result into Eq. (2) along with a new variable, $\zeta = z/k_0$, to obtain

$$i n_0^{(j)} \frac{\partial}{\partial \zeta} u = -\frac{1}{2} \frac{\partial^2}{\partial x^2} u - \frac{1}{2} \frac{\partial^2}{\partial y^2} u - 2\pi \frac{P}{P_{cj}} |u|^2 u . \tag{3}$$

Now let us consider the hypothetical situation in which two beams of light with identical normalized amplitudes $u(x, y)$ enter two different samples, which we denote by the superscripts $j = r$ (reference sample) and $j = t$ (test sample). We let the samples have linear indices of refraction $n_0^{(r)}$ and $n_0^{(t)}$ and thicknesses L_r and L_t. If the power is small enough that the last term in Eq. (3) can be neglected, and if the sample lengths are chosen so that $L_t/n_0^{(t)} = L_r/n_0^{(r)}$, it follows from Eq. (3) that the normalized amplitudes are identical at the exit faces of the two samples. Furthermore, the normalized amplitudes will be nearly identical at the exit faces of the two samples if $|L_t/n_0^{(t)} - L_r/n_0^{(r)}| \ll z_{d0}$, where z_{d0} is the Rayleigh range[11] in free space. If the input power is increased to some large values P_r and P_t, and if the nonlinear indices of refraction of the samples are $n_2^{(r)}$ and $n_2^{(t)}$, we see from Eq. (3) that to obtain the same $u(x, y)$ at the exit faces of the two samples, we should adjust the powers so that $[L_t/n_0^{(t)}](P_t/P_{ct}) = [L_r/n_0^{(r)}](P_r/P_{cr})$. For two samples of the same thickness $L_t = L_r = L$, this condition is equivalent to $P_t n_2^{(t)} = P_r n_2^{(r)}$. With the sample thicknesses properly selected and the powers properly adjusted, $u(x, y)$ will be the same for both samples at any given distance from the exit faces, and therefore the measured normalized peak-to-valley transmittances $\Delta T_{pvj} = [P_{pj}^{(det)} - P_{vj}^{(det)}]/P_{jave}^{(det)}$ will also be the same. Here $P_{pj}^{(det)}$ and $P_{vj}^{(det)}$ are the maximum (peak) and minimum (valley) powers that are registered for the jth sample of the detector (det) after it passes through the aperture. The average or baseline power is $P_{jave}^{(det)} = [P_{pj}^{(det)} + P_{vj}^{(det)}]/2$.

Following this analysis, we see that a simple procedure for making a Z-scan measurement is as follows: (1) Obtain reference and test samples of equal thicknesses L for which $|L/n_0^{(t)} - L/n_0^{(r)}| \ll z_{d0}$. (2) Make a Z-scan measurement of one of the samples. The exact size and shape of the aperture do not matter. For example, an obscuration disk (as in an eclipsing Z scan[12]) can be used. (3) Insert the second sample and adjust the input power until the normalized peak-to-valley transmittance ΔT_{pvj} matches that obtained for the first sample. (4) Calculate the nonlinear index of refraction using the following formula:

$$n_2^{(t)} = n_2^{(r)} P_r / P_t . \tag{4}$$

For a thin sample, it is not necessary to match the lengths as indicated in step (1) above, since the beam does not evolve appreciably (in either size or shape) in traversing the sample. For the special case in which the nonlinear phase shift is much less than unity, step (3) may also be simplified. To see how, we first note that $I(x, y, t; z) = P(t)|u(x, y; z)|^2$. The nonlinear phase shift for a thin sample can then be written as $\Delta\phi_j(x, y, t; z) = \omega_0 n_2^{(j)} L_j P_j(t) |u(x, y; z)|^2/c$. If $\Delta\phi_j \ll 1$, and if the electric-field amplitude at the entrance face of the sample is $A_1(x, y, t; z) = \sqrt{N(t)}\, u_1(x, y; z)$, then the amplitude at the exit face of the sample is $\sqrt{N}\, u_1 \exp(i\Delta\phi_j) \approx \sqrt{N}(u_1 + i\Delta\phi_j u_1) = \sqrt{N}(u_1 + \gamma_j B_1)$, where $\gamma_j = n_2^{(j)} L_j P_j(t)$ and $B_1 = i\omega_0 |u_1|^2 u_1/c$. For the experimental procedure considered here, if the entrance faces of the test and reference samples are located the same distance from (and on the same side of) the beam waist, then both samples will have the same values for u_1 and B_1 but different values for γ_j. The electric-field amplitude after propagation to the aperture can be written as $\sqrt{N}(u_2 + \gamma_j B_2)$, where u_2 and B_2 are again identical for the test and reference samples. The normalized transmittance (for a sample located at some arbitrary position) is defined as $\Delta T_j = [P_j^{(det)} - P_{jave}^{(det)}]/P_{jave}^{(det)}$. We can evaluate this quantity by using $\Delta T_j = (\iiint dx dy dt |u_2 + \gamma_2 B_2|^2 / \iiint dx dy dt |u_2|^2) - 1 \approx 2\gamma_j \,\mathrm{Re} \iiint dx dy dt\, u_2 B_2$. In these expressions, the integrations over x and y are performed over the extent of the aperture; the integration over t represents the action of the detector. Since the quantity $\mathrm{Re} \iiint dx dy dt\, u_2 B_2$ is the same for the test and reference samples, it follows that $\Delta T_r/\gamma_r = \Delta T_t/\gamma_t$ and from this that $\Delta T_{pvr}/\gamma_r = \Delta T_{pvt}/\gamma_t$. Substituting into this equation the expressions for γ_r and γ_t, we get

$$n_2^{(t)} = n_2^{(r)} \frac{\Delta T_{pvt} L_r P_r}{\Delta T_{pvr} L_t P_t} . \tag{5}$$

When applicable, this formula permits a simplification of the measurement procedure since the power can be set to any convenient value. In other words,

Table 1. Ratio of n_2 Values for Two Pairs of Liquids as Measured at $\lambda_0 = 1064$ nm with Five Cuvette Thicknesses

Cuvette Thickness (mm)	n_2(toluene)/ n_2(glycerine)	n_2(methanol)/ n_2(water)
1	14.1	1.05
2	14.6	1.07
5	14.4	1.06
10	14.2	1.07
20	14.0	1.07
Average	14.3	1.06

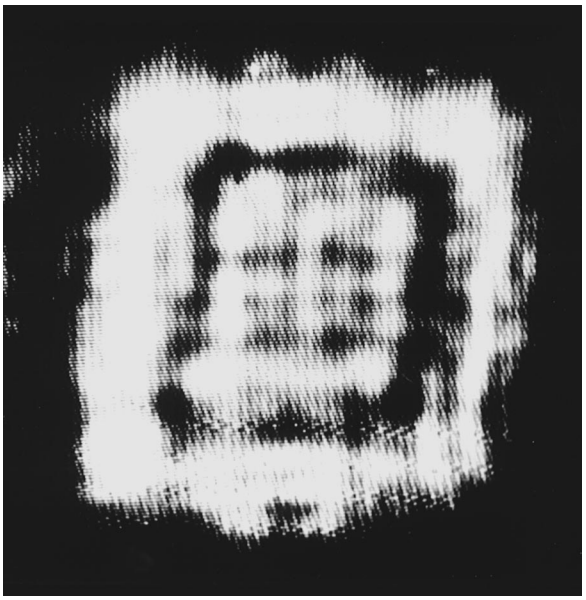

Fig. 1. Aberrated beam obtained by sending light from a Nd:YAG laser through a distorted wire mesh.

it is not necessary to perform the adjustment of step (3) given above. Furthermore, if $n_2^{(t)} \ll n_2^{(r)}$, we can use $P_t \gg P_r$, thereby avoiding damage to the reference sample. Equation (5) is applicable if the sample thicknesses are much less than a Rayleigh range and if the nonlinear phase shift is much less than unity. One can check that this latter condition is satisfied by verifying that $\Delta T_{\mathrm{pv}t} \propto P_t$ and $\Delta T_{\mathrm{pv}r} \propto P_r$.

To demonstrate the validity of the techniques described in this Letter, we carried out two experiments. In the first experiment, we measured the relative values of n_2 for two pairs of liquids: glycerine versus toluene and water versus methanol. We did this by following the procedure described in the paragraph preceding Eq. (4). The experiment was carried out using a Nd:YAG laser operating at 1064 nm and producing pulses of approximately 30-ps duration. The laser light was sent through a half-wave plate, a polarizer, a neutral-density filter (when necessary), and then through the usual elements of a Z-scan system—a lens, a sample under test (near the beam waist), an aperture (in the far field), and a detector. The half-wave plate was used to adjust the power at the entrance face of the sample. As shown in Table 1, we used cuvette thicknesses of 1, 2, 5, 10, and 20 mm. The two largest thicknesses exceeded the Rayleigh range of our experimental arrangement, which was somewhat less than 1 cm. For glycerine and toluene, the n_0 values are 1.47 and 1.48, respectively, and for water and methanol, they are 1.33 and 1.32, so for both pairs the condition $|L/n_0^{(t)} - L/n_0^{(r)}| \ll z_{\mathrm{d}0}$ is easily satisfied. Table 1 shows that consistent results were obtained for all five sample thicknesses for both pairs of liquids. The n_2 of toluene was found to be ≈14.3 times that of glycerine, and the n_2 of methanol was found to be ≈1.06 times that of water.

In the second experiment, we used the same procedure as in the first experiment to measure n_2 of toluene ($n_0 = 1.48$) relative to acetone ($n_0 = 1.35$), using a 10-mm cuvette thickness. [In this case, $|L/n_0^{(t)} - L/n_0^{(r)}| \approx z_{\mathrm{d}0}/10$, which is acceptable.] This time, however, we first used the beam from the Nd:YAG laser without alteration and next used the beam shown in Fig. 1, which we obtained by sending the light from the laser through a distorted wire mesh. We obtained good agreement using the two different beams. For the beam directly from the laser, we found $n_2(\mathrm{toluene})/n_2(\mathrm{acetone}) = 6.32$; for the aberrated beam, we found $n_2(\mathrm{toluene})/n_2(\mathrm{acetone}) = 6.41$.

In conclusion, we have presented a new Z-scan measurement technique that accurately determines the value of n_2 for a test sample relative to that of a reference sample. The technique is easy to carry out. It permits the use of a non-Gaussian beams and either thick or thin samples, and it eliminates the need to know the details of the pulse shape.

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