

# APPLICATION NOTE

## Z-Scan for the Characterization of Transparent Optical Materials

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## Introduction

Reliable methods for determining the nonlinear optical properties of materials (i.e. nonlinear absorption and nonlinear refraction) have been developed for wide ranging applications such as optical limiting<sup>1</sup>, multi-photon polymerization<sup>2</sup> as well as optical switching<sup>3</sup>. Of these methods, “z-scan”, developed by Eric Van Stryland<sup>4</sup>, remains the standard technique.

The z-scan technique is performed by translating a sample through the beam waist of a focused beam and then measuring the power transmitted through the sample. Z-scan has many possible configurations (e.g. “EZ-Scan”, “White Light z-scan”, “Excite-Probe z-scan”). In this note, only the standard “open aperture” and “closed aperture” z-scan will be discussed. The two measurable quantities connected with the z-scan are nonlinear absorption (NLA) and nonlinear refraction (NLR). These parameters are associated with the imaginary and real part of the third order nonlinear susceptibility,  $\chi^{(3)}$ , and provide important information about the properties of the material.

The purpose of this note is to describe a simple implementation of the z-scan technique that can be used to characterize relatively thin (< 5mm) optical materials.

## Theory of Operation

Figure 2.1 illustrates the z-scan apparatus described in this note. An intense laser beam is sent through a “long” focal length lens. A “thin” sample is translated through the beam waist using a motorized translation stage. The beam after focus is split by means of a beamsplitter, and a fraction of the beam is sent to a photodiode (Photodiode 1) where the beam under-fills the active area of the diode. The remainder of the beam is sent through an aperture (iris) which clips roughly

half of the beam intensity. After the aperture, a second photodiode (Photodiode 2) detects the remainder of the beam passing through the iris. The output of both photodiodes is sent to a dual channel power meter interfaced to a PC. In this configuration, both the “closed” and “open” aperture z-scans can be simultaneously obtained using only one measurement.

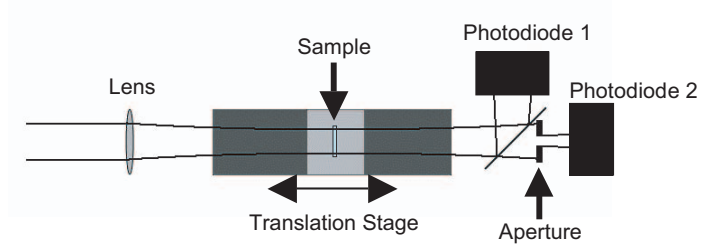


Figure 2.1: Illustration of the experimental setup for z-scan.

It is worth noting at this point that both detectors can be replaced with a single CCD camera. The open aperture result can be obtained by integrating over the entire CCD array. Eliminating the need for an aperture prior to the detector, analogous information to the closed aperture result can be obtained by determining the change in the radius of the beam as the sample is translated through the focal region of the beam. In fact, if the beam size at the focusing lens is well known, the  $M^2$  value can be extracted since the z-scan effectively measures the Rayleigh range in addition to the nonlinear properties of the material.

Adhering to the convention of Sheik-Bahane et al.<sup>4</sup>, the following notations will be used:

$\alpha$	= absorption coefficient
$\beta$	= two-photon absorption coefficient
$\lambda$	= laser wavelength
$\Delta\phi_o$	= on-axis peak nonlinear phase shift with sample at focus
$\Delta T(Z)$	= normalized transmittance of the sample at Z
$\Delta T_{pv}$	= change in normalized transmittance between peak and valley
$\chi^{(3)}$	= third order susceptibility
I	= Irradiance (power per unit area)
$I_o$	= peak on-axis irradiance at focus
L	= sample length
$L_{eff}$	= $(1 - e^{-\alpha L})/\alpha$
n	= index of refraction
$q_o$	= $\beta I_o L_{eff}$
S	= fraction of beam transmitted by the aperture
t	= time
T(Z)	= transmittance of the sample at Z
$w_o$	= spot size at focus (radius at $1/e^2$ )
Z	= position of sample with respect to the focal position
$Z_o$	= Rayleigh range $n\pi w_o^2/\lambda$

In the above table,  $\Delta T(Z)$  and  $\Delta T_{pv}$ , referred to as normalized parameters, require clarification. Normalization in this context is defined as the transmittance of the sample, T(Z), divided by the transmittance of the sample far from focus (i.e.  $Z \gg 0$ ). Using this definition,  $\Delta T(Z)$  and  $\Delta T_{pv}$  reach a value of one far from focus and deviate from one as the sample is translated through the focus of the beam.

The simple expressions presented in the next two sections for data analysis make the following assumptions:

1. The pulse envelope evolves on a much longer time scale than the optical frequency (i.e. the slowly varying envelope approximation).<sup>4</sup> The ramification of this is that the following equations will not hold for few optical cycle pulses.
2. The length of the sample must be less than the Rayleigh range of the beam (i.e.  $L < Z_o$ ) and less than the Rayleigh range divided by the peak nonlinear phase shift (i.e.  $L < Z_o / \Delta\phi_o$ ). The second criteria is usually satisfied given the first criteria is met since the nonlinear phase shift is most often small.<sup>4</sup>
3. The beam profile, in both time and space, follows a Gaussian distribution.<sup>4</sup>
4. The nonlinear response is restricted to third order (i.e.  $\chi^{(3)}$  processes).<sup>4</sup>

### Open Aperture – Nonlinear Absorption

As the sample is translated through the focal region of the beam, Photodiode 1 measures the total transmitted intensity. Because only the irradiance at the sample is changing as the sample is translated, any deviation in the total transmitted intensity must be due to multi-photon absorption. In the limit where multi-photon effects are limited to two-photon absorption, the normalized change in transmitted intensity can be approximated by the following equation,

$$\Delta T(Z) \approx \frac{q_o}{2\sqrt{2}} \left[ \frac{1}{1 + Z^2 / Z_o^2} \right] \quad (1)$$

Figure 2.2 illustrates the normalized transmittance ( $\Delta T+1$ ) as a function of sample position ( $Z$ ) based on Equation 1 (the values for the Rayleigh range ( $Z_0$ ) and  $q_0$  used to generate this curve correspond to 18 mm and 0.5 respectively). Negative values for  $Z$  indicate that the sample is positioned between the focusing lens and the beam waist at focus, and positive values for  $Z$  indicate that the sample is between the beam waist at focus and the detector. Once the open aperture data is collected, it can be readily fit to Equation 1. The two-photon absorption coefficient,  $\beta$ , can be determined from the relation,

$$q_0 = \beta I_0 L_{\text{eff}} \quad (2)$$

For transparent samples,  $L_{\text{eff}} \approx L$  where  $I_0$  is simply the peak irradiance at the focus.

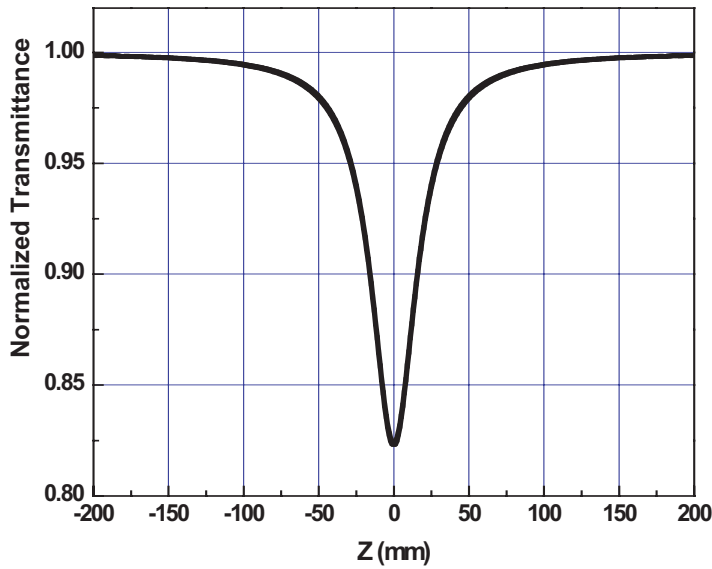


Figure 2.2: Example of nonlinear absorption in a z-scan experiment. The values for the Rayleigh range ( $Z_0$ ) and  $q_0$  used to generate this curve correspond to 18 mm and 0.5 respectively.

### Closed Aperture – Nonlinear Refraction

As the sample is translated through the focal region of the beam, Photodiode 2 measures the fraction of intensity passing through the aperture which is transmitted through the sample. If the beam experiences any nonlinear phase shift due to the sample as it is translated through the focal region, then the fraction of light falling on the detector will vary due to the Kerr lens generated in the material by the intense laser beam.

In this case, the signal measured by Photodiode 2 will exhibit a peak and valley as the sample is translated (see Figure 2.3). The position of the peak and valley, relative to the  $Z$ -axis, depends on the sign of the nonlinear phase shift,  $\Delta\phi_0$ .

If the phase shift is positive (i.e. self-focusing occurs), then the peak will trail the valley. If the phase shift is negative, then the valley will trail the peak (i.e. self-defocusing). The magnitude of the phase shift can be determined from the change in normalized transmittance between peak and valley,  $\Delta T_{pv}$ , using the relation,

$$|\Delta\Phi_0| = \frac{\Delta T_{pv}}{0.406(1-S)^{0.27}} \quad (3)$$

where  $S$  is the fraction of beam transmitted through the aperture. The nonlinear index,  $n_2$ , can then be determined from the phase shift using,

$$n_2 = \left( \frac{\lambda}{2\pi} \right) \Delta\Phi_0 / I_0 L_{\text{eff}} \quad (4)$$

The above relations are based on approximations which are accurate to within 3%. If further accuracy is required, a more precise method based on Gaussian Decomposition is presented by Sheik-Bahae et al<sup>4</sup>.

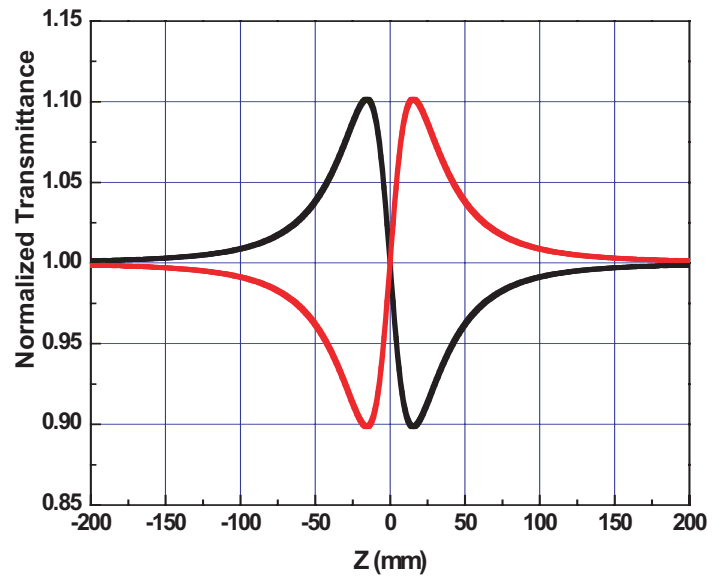


Figure 2.3: Example of nonlinear refraction in a z-scan experiment. The red curve corresponds to a positive phase shift (self-focusing) while the black curve corresponds to a negative phase shift (self-defocusing).

## Closed Aperture – Nonlinear Refraction in the Presence of Nonlinear Absorption

When both NLR and NLA are present in the same measurement, the closed aperture measurement will become asymmetric resembling that shown in Figure 2.4 A. In order to determine the nonlinear phase shift, the closed aperture scan can be divided by the open aperture scan (Figure 2.4 B). The result of this division, shown in Figure 2.4 C, can be used to estimate the closed aperture results in the absence of nonlinear absorption.<sup>4</sup>

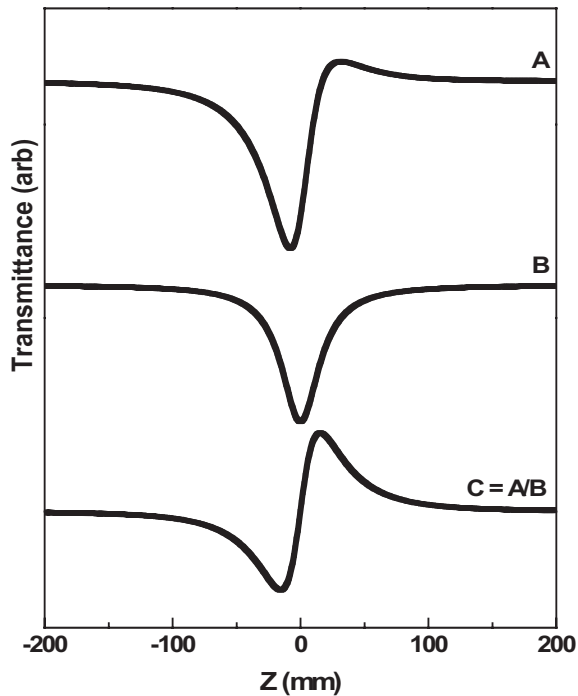


Figure 2.4: Simulated z-scan signal with both NLR and NLA present. The top curve (A) is the result of the closed aperture scan. The middle curve (B) is the result of the open aperture scan. The bottom curve (C) estimates the closed aperture result in the absence of NLA, and is obtained by dividing the actual closed aperture measurement (A) by the open aperture measurement (B).

## Experimental Setup

Figure 3.1 shows the experimental z-scan setup constructed in the Technology and Applications Center at Newport. The light source is the Spitfire® XP Pro (not shown), and consists of two routing mirrors, two irises, a 500 mm focal length lens, a translation stage, a beam splitter, two photodiodes and a two-channel power meter (specific part numbers are listed in the appendix).

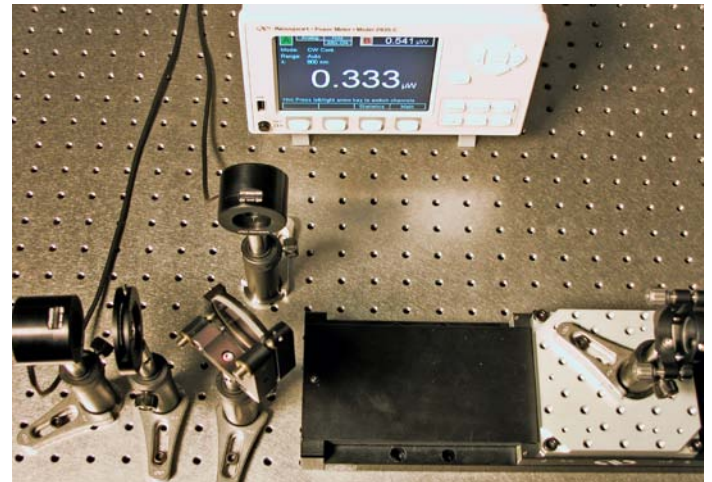
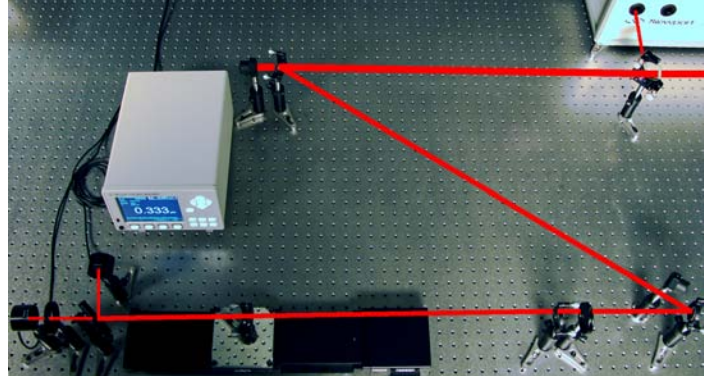


Figure 3.1: (Top) Full experimental setup for z-scan. (Bottom) Expanded view of the detection system.

The beam emerging from the Spitfire XP (3 mJ) at 1kHz, 35 fs FWHM) is aligned parallel to the axis of translation of the stage using the two routing mirrors (Note, the routing mirrors may be replaced with beam samplers in order to attenuate the pulse energy, if necessary). An iris is placed after the second routing mirror in order to clean up the edges of the beam. The 500 mm focal length lens is placed after the iris leaving some room for neutral density filters, if needed. The translation stage is positioned such that the center of translation corresponds to the location of the beam waist at the focus. The sample, for this application either a 3mm window of BK7 or fused silica, is inserted into the beam path such that it is also located in tight focus. A second iris is positioned in the beam path after the stage so that its distance from the sample is significantly greater than the Rayleigh length of the focused beam. Photodiode 2 is then placed immediately after this iris. The iris should be closed to aperture the beam. Transmitted power readings are noted with the iris open and closed (the ratio of the transmitted power with the iris open and closed provides the value for  $S$  used in Equation 3).

Between the translation stage and second iris, a beamsplitter is inserted to pick off a fraction of the beam. Photodiode 1 is carefully placed after the beamsplitter, so as not to overfill the detector. The fraction of the beam picked off by the beamsplitter is noted (approximately 20% for P polarization and 50% for S polarization).

The two photodiodes are connected to the power meter which is interfaced to a PC. For this set of experiments, data is collected using an in-house LabView program. For the experiments shown below, the beam is further attenuated using reflective neutral density filters placed between the iris and the 500 mm focal length lens.

Figure 3.2 shows the z-scan data collected using this setup for both a 3 mm BK7 window and a 3 mm Fused Silica window. The black curves correspond to the open aperture measurements, while the red curves correspond to the closed aperture measurements. Analysis of the closed aperture scans using Equations 3 and 4 yield values for  $n_2$  of  $2.9 \cdot 10^{-20} \text{ m}^2/\text{W}$  for fused silica and  $9.7 \cdot 10^{-20} \text{ m}^2/\text{W}$  for BK7. Both of these values agree within a factor of two with that reported in the literature.<sup>8</sup> BK7 exhibits a value of  $n_2$  roughly three times greater than that of fused silica; moreover, BK7 exhibits nonlinear absorption at the peak power used for these experiments (experimental parameters:  $I_0 = 6 \cdot 10^{14} \text{ W}/\text{m}^2$ ,  $Z_0 = 18 \text{ mm}$ ). It is worth noting that these peak powers are accessible in the unfocused output of the Spitfire<sup>®</sup> XP as well as many other commercial regenerative amplifiers. For this reason, optics fabricated with BK7 should not be used with such high intensity systems, unless the intensity is first reduced by splitting the beam or expanding it.

### Conclusion

We describe a setup for measuring the nonlinear optical properties of materials based on the z-scan technique. This implementation is a simple method for determining the nonlinear absorption and nonlinear refraction of optical materials. The peak powers needed to drive these processes are achievable from the direct output of the Spitfire XP as well as several other commercial regenerative amplifiers. These measurements provide valuable information that can be used to determine which optical materials are suitable for use in a particular experimental setup. In some cases, reflective optics may be the only choice with high intensity ultrashort laser pulses.

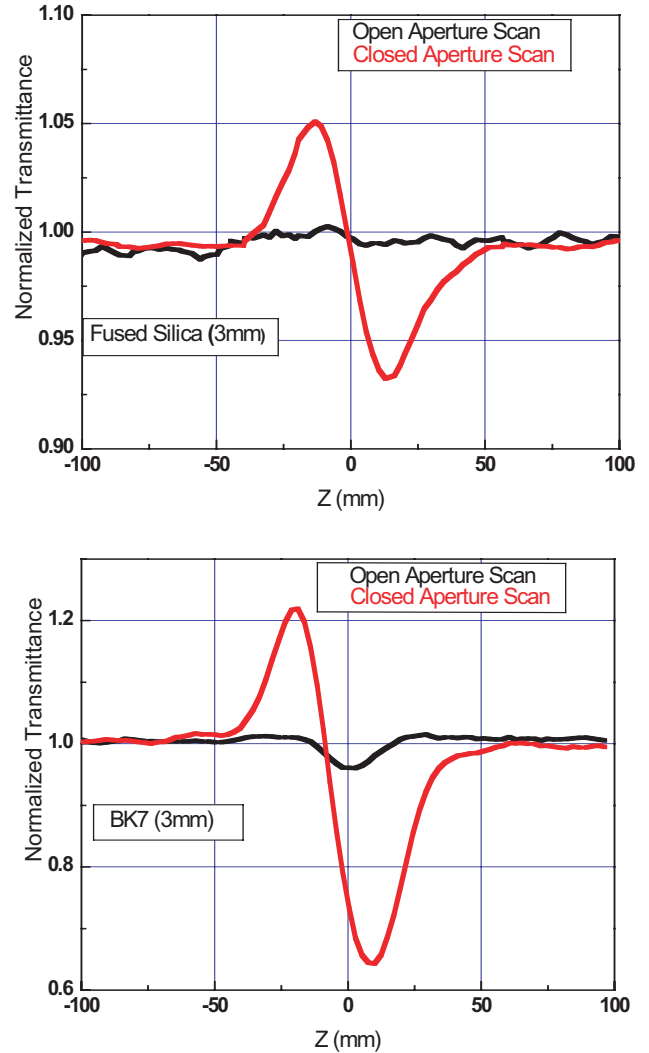


Figure 3.2: Open (black) and closed (red) aperture results for the z-scan measurements for fused silica (top) and BK7 (bottom).

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## Appendix 1. Running the Software

Prior to connecting any device, install the SMC100CC and power meter software according to the procedure specified on the installation disk. Reboot the computer. Connect the computer through the USB port located on the rear of the power meter. Connect the SMC100CC controller to the computer using the RS232 cable provided. Run the SMC100CC program from the Start Menu. Make sure the stage is properly configured and sent to the home position. Command the stage to its center position (125 mm). Note the COM port address. Close the SMC100CC program. After successful completion of this step, a green LED should be illuminated on the SMC100CC controller. Connect the two photodiodes to the rear of the power meter.

The software provided (see Figure A1.1) is non-compiled source code written in Labview 8.2. Start Labview 8.2 and open the z-scan code. Initialize the code by pressing the white arrow button at the top-left corner of the screen. After the code is initialized, enter values for the Home Position (125 mm = center of travel), the number of steps, the number of averages and the COM port (the same as above). After the appropriate values have been entered, press the START button. The system will begin acquiring data and moving the translation stage. When the run is finished, a dialog box will pop-up prompting the user to save the data. The user can enter a filepath, name and extension for the file (if the user does not wish to save the data, press the CANCEL button). The data is saved as a series of three columns: The first column corresponds to stage position, the second to the open aperture measurement and the third to the closed aperture measurement.

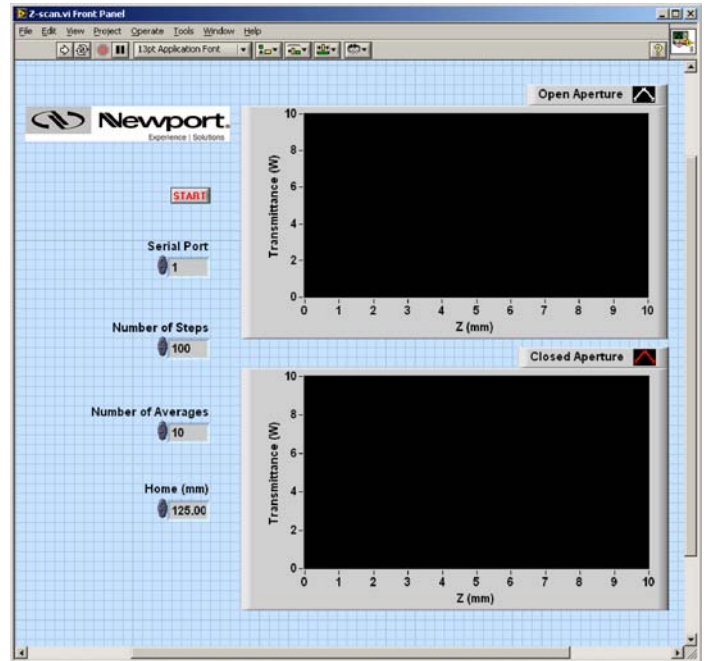


Figure A1.1: Example z-scan software



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