# Z-Scan Beam Simulation

## 1.1. Z-Scan Technique

Nonlinear optical material properties can be characterized using the Z-Scan measurement technique. The Z-Scan determines the NLA and NLR of a material by scanning it through the focus of a high-power ultrafast laser source. As the sample approaches focus, the irradiance and hence the induced nonlinear effects increase. By measuring the energy received at the closed and open apertures as a function of sample position relative to the focus, the NLR and NLA can be determined.

There are two signals produced by the experiment. The open aperture (OA) signal is due to the change in absorption as the sample approaches the focus. For $\alpha_2 > 0$ this yields a reduction in the transmission near focus as seen in Figure 1.

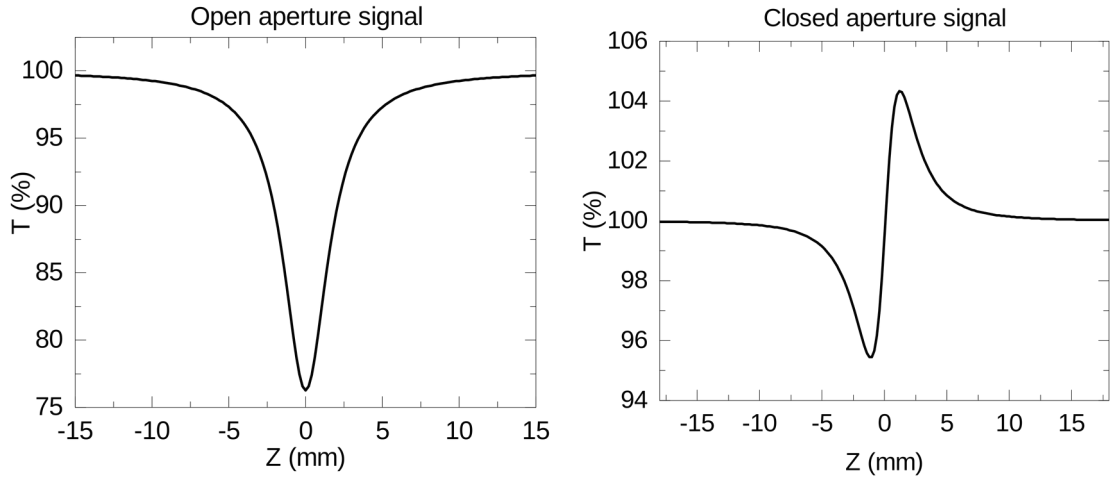


Figure 1: Left) Open aperture signal. For $\alpha_2 > 0$ the absorption increases near focus, resulting in a reduction in the transmission. Right) Closed aperture signal. For $n_2 > 0$ the self-focusing of the beam before and after the focus causes the spot size to change on the aperture, thus yielding the familiar valley-peak structure in the transmission.

The signal from the closed aperture (CA) is shown in Figure 1. As the sample approaches the focal point, the self-focusing of the beam causes the beam to focus to a tighter spot, which in turn causes the divergence angle of the beam to also increase. This causes the spot size of the beam on the aperture to increase, which in turn reduces the transmission. On the other side of focus, this self-focusing causes the spot size to be smaller on the aperture, thus increasing the transmission as seen in Figure 2. In the case where we have both NLR and NLA, the CA signal contains influence from both effects.

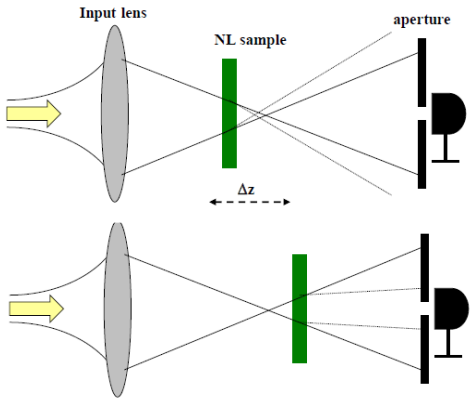


Figure 2: Closed aperture Z-Scan geometry. The nonlinear self-focusing makes the spot size on the aperture smaller as the sample approaches focus, while causing the spot size to expand on the other side of focus, thus altering the transmission through the aperture.

## 1.2. Z-Scan Fitting Code

Once the data is collected it needs to be fit to extract the nonlinear values. The fitting code is based on a quasi-analytical analysis. In this approach we analytically solve for the field at the back of the sample in the slowly varying envelope, thin sample ($L_{eff} \ll z_0$) and small nonlinear phase change ($\Delta\phi_0 = k_0 n_2 I_0 \;\; L_{eff} \ll 1$) approximations where $L_{eff}$ is the effective sample thickness, $z_0$ is the Rayleigh range, $k_0$ is the free space wave number and $I_0$ is the peak irradiance. We then Fresnel propagate the electric field to the aperture plane and calculate the power passing through the open and closed apertures. The signals are then normalized by their values far from focus ($Z \gg 0$) to give the normalized transmission $T$ as a function of sample position $Z$.

We start with the assumption that nonlinearity is instantaneous (absorption change $\Delta\alpha = \alpha_2 I$ and index change $\Delta n = n_2 I$). The governing equations for the propagation are

$$\frac{dI}{dz'} = -\alpha_0 I - \alpha_2 I^2 \tag{2}$$

$$\frac{d\phi}{dz'} = k_0 n_2 I \tag{3}$$

where $z'$ is the coordinate the sample frame, and $\phi$ is the phase. The input Gaussian beam $E_{in}[Z, \rho, T]$ is given by

$$R[Z] = \frac{1}{Z} + Z \tag{4}$$

$$W[Z] = \sqrt{1 + Z^2} \tag{5}$$

$$E_0[T] = \exp\left[-\frac{1}{2}T^2\right] \tag{6}$$

$$E_{\text{in}}[Z,\rho,T]=\frac{E_0[T]}{W[Z]}\exp\left[-\frac{\rho^2}{W[Z]^2}-I\frac{\rho^2}{R[Z]}\right] \tag{7}$$

where in the normalized coordinate system is $Z=z/z_0$, $\rho=r/w_0$ and $T=t/\tau$, where $w_0$ is the beam waist (HW $1/e^2$ of the irradiance), $\tau$ is the pulse duration (HW $1/e$ of the irradiance) and $r$ is the radial distance. $R[Z]$ is the phase front radius of curvature, $W[Z]$ is the spot size and $E_0[T]$ is the temporal part of the field.

We analytically solve equations 2 and 3 for the electric field at the back of the sample. The field at the back of the sample $E_e[Z,\rho,T]$ is

$$q[Z,\rho,T]=\alpha_2 I_{\text{in}}[Z,\rho,T]L_{eff} \tag{8}$$

$$E_e[Z,\rho,T]=E_{\text{in}}[Z,\rho,T]e^{-\frac{\alpha L}{2}}\left(1+q[Z,\rho,T]\right)^{\left(\frac{ik n_2}{\alpha_2}-\frac{1}{2}\right)} \tag{9}$$

where $q[Z,\rho,T]$ is the dimensionless nonlinear absorption parameter, $I_{\text{in}}[Z,\rho,T]=\frac{1}{2}nc\epsilon_0\left|E_{\text{in}}[Z,\rho,T]\right|^2$ is the input irradiance and $L_{eff}=\left(1-\exp\left[-\alpha_0 L\right]\right)/\alpha_0$ where $L$ is the sample length. We then need to numerically propagate this to the aperture plane using the Fresnel transform approach. The field at the aperture plane is

$$E_a[Z,\rho,T]=F^{-1}\left\{F\left\{E_e[Z,\rho,T]\right\}\cdot F\left\{\frac{\exp[ikz]}{ikz}\exp\left[i\frac{k}{2z}\rho^2\right]\right\}\right\} \tag{10}$$

where $F$ indicates the Fourier transform and $F^{-1}$ is the inverse Fourier transform. This must be performed numerically and is the most computationally intensive part of the calculation. Now that we have the electric field at the aperture plane, we integrate numerically over space and time to get the OA signal. The normalized transmission $T_{OA}$ is calculated by normalizing the OA signal by its value far from focus.

$$OA[Z]=\int_{-\infty}^{\infty}\int_{0}^{\infty}\left|E_a[Z,\rho,t]\right|^2 2\pi\rho\ \ d\rho\, dT \tag{11}$$

$$T_{OA}[Z]=\frac{OA[Z]}{OA[Z\ll 0]} \tag{12}$$

A similar procedure is performed for the CA signal, except we integrate only to the closed aperture radius $\rho_{max}$ and then normalize to calculate $T_{CA}$.

$$CA[Z]=\int_{-\infty}^{\infty}\int_{0}^{\rho_{max}}\left|E_a[Z,\rho,t]\right|^2 2\pi\rho\ \ d\rho\, dT \tag{13}$$

$$T_{CA}[Z]=\frac{CA[Z]}{CA[Z\ll 0]} \tag{14}$$

This code has the advantage over other approximation methods in that it can handle larger phase changes than the Gaussian decomposition method (usually limited to $\Delta T<10\,\%$) as

well as accounting for NLA. It is limited in that it assumes that the nonlinearities are instantaneous and thus cannot account for carrier redistribution, molecular rotation, and other non-instantaneous nonlinearities.

The code first uses the reference data to remove the noise due to laser power fluctuations. Next the code accounts for the linear background by fitting a line across the data and removing the linear slope, which also normalizes the transmission. An example of this window is shown in Figure 3.

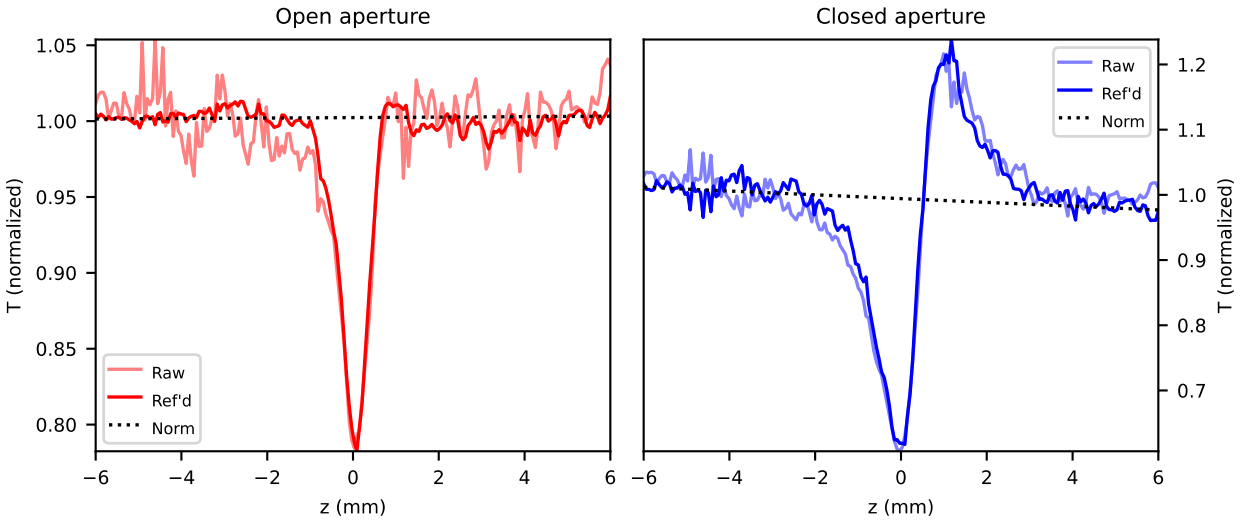


Figure 3: Reference processing and background removal window. The reference data is used to remove the noise (thin and thick lines), while the linear background (black line) is removed from the signal.

Once this is completed the theoretical OA and CA signals are calculated and displayed along the data as seen in Figure 4. The user can then manually adjusts the nonlinear parameters until the theory and data match up. The values can be further refined using the downhill simplex function which will minimize the RMSE for the selected parameters. It is recommended that the minimum number or parameters be optimized at once, preferably one if possible. Once the fitting is complete, the data and fitting parameters can be exported. The code outputs four files 1) a comma separate file containing the data and fit values at each stage position, 2) a settings file listing all setting used in the fitting of the data, 3) an image file of the fit in JPG format and 4) and image of the fit in SVG format.

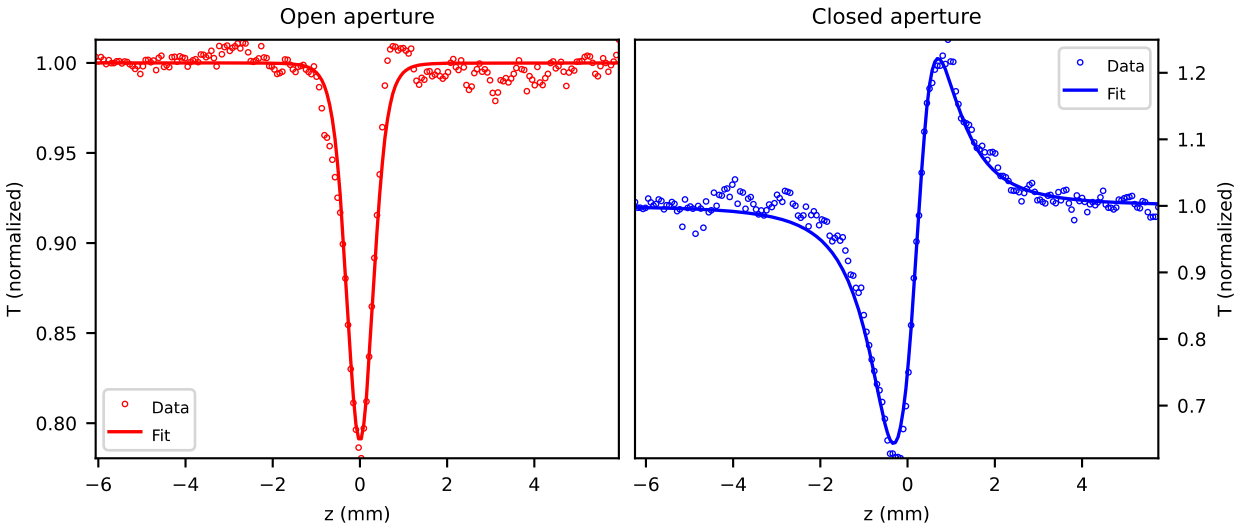


Figure 4: OA and CA fitting windows. The normalized transmission data (open circles) and the theoretical fit lines (solid) are displayed together allowing for manual fitting of nonlinear parameters.

## 1.3. Fitting Code Parameters

The processing options and fitting parameters for the Z-Scan fitting code are given in Table 1 and Table 2.

Table 1: Processing options for Z-Scan fitting code.

| **Z-Scan Fitting Code Options** | | |
|---|---|---|
| Parameter | Type | Purpose |
| Data file | Text | Location of data file to import. |
| Delimiter | Text | Delimiter that separates data columns in the data. If blank defaults to tab-delimited. |
| Extra label | Text | Additional label to append to output data files. Useful of performing multiple fits on the same data. |
| z column | Integer | Column containing the sample position. |
| CA column | Integer | Column containing the closed aperture data. |
| OA column | Integer | Column containing the open aperture data. |
| REF column | Integer | Column containing the reference detector data for power normalization. |
| FREQ column | Integer | Column containing the demodulation frequency data from lock-in amplifiers. |
| Use reference channel | Boolean | Use the reference data to remove noise. |
| Use frequency channel | Boolean | Use the frequency data to remove noise. |
| Flip z | Boolean | Reverse the sample position axis. |
| Normalize OA | Boolean | Normalize the open aperture signal using linear fitting. |
| Normalize CA | Boolean | Normalize the closed aperture signal using linear fitting. |
| Flip OA | Boolean | Flip the signal about unity. |

| | | |
|---|---|---|
| Flip CA | Boolean | Flip the closed aperture signal about unity. |
| Flip REF | Boolean | Flip reference signal about unity. |
| Flip OA display | Boolean | Flip open aperture signal in fitting display. |
| Flip CA display | Boolean | Flip closed aperture signal in fitting display. |
| Show CA | Boolean | Show closed aperture closed over open aperture signal in the fitting display. |
| z points | Integer | Number of z-points to calculate the signals. The code uses spline interpolation to calculate the value of the signals at each z data point. |
| r points | Integer | Number of points to calculate the field along the radial direction. |
| r max | Integer | The range over which to calculate the field in terms of spot size. |
| t points | Integer | Number of points to calculate the field along the temporal direction. |
| t max | Integer | The range over which to calculate the field in terms of pulse duration. |
| $r_a$ points | Integer | The number of point to sample the field at the closed aperture in the radial direction. |

Table 2: Fitting parameters for Z-Scan fitting code.

| **Z-Scan Fitting Code Parameters** | | |
|---|---|---|
| Parameter | Units | Purpose |
| Nonlinear refraction $n_2$ | $m^2/W$ | Second order nonlinear refraction coefficient. |
| Nonlinear absorption $\alpha_2$ | $m/W$ | Second order nonlinear absorption coefficient. |
| OA shift $\Delta_{OA}$ | None | Shift of theoretical open aperture signal curve. |
| CA shift $\Delta_{CA}$ | None | Shift of theoretical closed aperture signal curve. |
| OACA shift $\Delta_{CAOA}$ | None | Shift of theoretical closed over open aperture signal curve. |
| Linear index $n$ | None | Surface reflection calculation. |
| Sample thickness $L$ | $m$ | Effective sample length calculation. |
| Linear transmission $T_{lin}$ | None | Effective sample length calculation. |
| Free space wavelength $\lambda$ | $m$ | Phase accumulation calculation. |
| Pulse energy $E$ | $J$ | Irradiance calculation. |
| Spot size $w_0$ | $m$ | Irradiance calculation. |
| Pulse duration $\tau_{FWHM}$ | $s$ | Irradiance calculation. |
| Beam parameter product $M^2$ | None | Irradiance calculation. |
| CA transmission $S$ | None | Transmission of closed aperture, usually set to 0.33. |
| CA distance $d$ | $m$ | Distance to closed aperture, usually set to 1 m. |

## 1.4. Credits

Initial algorithm developed by Matthew Riechert.
Subsequent development and GUI developed by Manuel Ferdinandus