On the measurement of high-order refractive nonlinearities through the Z-scan technique

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Abstract. High-order refractive nonlinearities play an increasingly important role for the characterization of new materials in relation to applications as optical switching, optical limiting, optical data processing, etc. In this work a novel more accurate method for determining the coefficients related to the higher-order refractive nonlinearities through the Z-scan technique is presented. The general case of an elliptic Gaussian incident beam is considered. The theoretical analysis is based on the Gaussian decomposition method, appropriately extended to higher-order nonlinearities, which are treated as a perturbation. An analytic expression for the electric field pattern of the beam at any distance \(D\) from the exit plane of the sample is obtained. This allows the analysis and simulation of Z-scan experiments based on either the measurement of the transmittance through an aperture or the direct measurement of the beam dimensions in the far-field, which has considerable advantages, especially in the case of elliptic beams. For both cases, an experimental procedure for measuring the high-order refractive nonlinearities is suggested.

1. Introduction

As the optical intensities involved in photonic experiments and applications become stronger the effect of higher-order nonlinearities becomes more significant. Especially, higher-order refractive nonlinearities play an increasingly important role in applications as optical switching, optical limiting, optical data processing, etc. Consequently, the new materials developed for use in these applications must also be characterized in regard to their higher-order nonlinear optical properties.

A simple and accurate method for measuring optical nonlinearities is the well-known z – scan technique [1], based on moving a thin sample of the material along the propagation direction \(z\) inside the focal region of a focused laser beam. The nonlinear refractive index of the material induces a phase shift across the profile of the laser beam, which causes variations to the far-field pattern of the beam as the sample moves inside the focal region. These variations provide information for both the sign and the magnitude of the refractive nonlinearity and they can be recorded by measuring either the transmittance of the irradiance through an aperture or directly the beam dimensions in the far field, as it has been recently proposed [2, 3]. It should be noted that the direct measurement of the beam dimensions provides several advantages [2], especially in the case of an elliptic Gaussian incident beam [3] where there is no symmetry between the beam and the aperture shape.

Theoretically, the far field pattern of the beam is obtained either numerically using the Huygen’s integral [4], or analytically through the Gaussian decomposition method [1]. According to this

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approach the electric field pattern at the exit plane of the sample is decomposed to a linear combination of Gaussian beams with decreasing waist dimensions through a Taylor expansion of the induced phase shift. Each individual Gaussian beam is easily propagated to any desired distance $D$ from the exit plane of the sample, and finally they are summed to reconstruct the electric field pattern of the laser beam at this distance.

In the present work the Gaussian decomposition method is extended to the case that the first two orders of refractive nonlinearity are simultaneously excited. The general case of an elliptic Gaussian beam is considered. Using the realistic assumption that the magnitude of the higher-order nonlinearity is much smaller than the magnitude of the lower-order one an analytic expression for the electric field pattern of the beam at any distance $D$ from the exit plane of the sample is obtained. Based on these results simulations of Z-scan experiments based on either the measurement of the transmittance through an aperture or the direct measurement of the beam dimensions in the far field are performed. For both cases an experimental procedure for measuring the higher-order nonlinearities is suggested.

2. Theoretical analysis
First we consider an astigmatic elliptic Gaussian beam, the electric field of which is given by the relationship \[ E(x, y, z, t) = E_0(t) \left( \frac{w_{0x} w_{0y}}{w_x(z) w_y(z)} \right)^{1/2} \exp \left( -\frac{x^2}{w_x^2(z)} - \frac{y^2}{w_y^2(z)} \right) \times \exp \left( -\frac{ikx^2}{2R_x(z)} - \frac{iky^2}{2R_y(z)} \right) \exp[-ikz + i\theta(z)] \] where $E_0(t)$ is a constant in space containing the temporal envelope of the electric field, $k$ is the wave number and $w_{0x}, w_{0y}$ the minor semiaxes at the two waists of the elliptic beam. The remaining parameters are the principal semiaxes $w_{x,y}(z)$, radii of curvature $R_{x,y}(z)$ and on-axis phase shift $\theta(z)$ of the beam at an arbitrary position $z$. They are given by the relationships \[ \begin{align*}
  w_{x,y}(z) &= w_{0x,y} \left[ 1 + \left( \frac{z - z_{0x,y}}{z_{R_{x,y}}} \right)^2 \right]^{1/2} \\
  R_{x,y}(z) &= \left( z - z_{0x,y} \right) \left[ 1 + \left( \frac{z_{R_{x,y}}}{z - z_{0x,y}} \right)^2 \right] \\
  \theta(z) &= \frac{1}{2} \tan^{-1} \left( \frac{z - z_{0x}}{z_{R_{x,y}}} \right) + \frac{1}{2} \tan^{-1} \left( \frac{z - z_{0y}}{z_{R_{y}}} \right)
\end{align*} \] where $z_{0x}, z_{0y}$, are the locations of the beam waists and $z_{R_{x,y}} = kw_{0x,y}^2/2$ the Rayleigh lengths for each principal direction $(x, y)$.

Next, let us suppose that this astigmatic elliptic beam is incident on a thin sample of a material characterized by both the first two orders of refractive nonlinearity. Namely, the refractive index $n$ of the material is given by the relationship $n = n_0 + n_2^{(3)} I + n_2^{(5)} I^2$ where $n_0$ is the linear refractive index of the material, $n_2^{(3)}$ and $n_2^{(5)}$ the third- and fifth-order nonlinear refractive indeces respectively, and $I$ the intensity of the incident beam. The effect of the refractive nonlinearities is to induce a phase shift of the form $\Delta \phi(x, y, z_\delta, t) = -kn_2^{(3)} I(x, y, z_\delta, t) L_{\text{eff}}^{(3)} - kn_2^{(5)} I^2(x, y, z_\delta, t) L_{\text{eff}}^{(5)}$ where $z_\delta$ is the location of
the sample and \( L_{\text{eff}}^{(3)} = \frac{1 - \exp(-a_0 L)}{a_0} \), \( L_{\text{eff}}^{(5)} = \frac{1 - \exp(-2a_0 L)}{2a_0} \) are the effective lengths corresponding to the third- and fifth-order nonlinearities respectively. Here \( L \) is the physical length of the sample and \( a_0 \) the linear absorption coefficients.

Consequently the electric field pattern \( E_e(x, y, z_s, t) \) of the beam at the exit plane of the sample becomes \( E_e(x, y, z_s, t) = E_{in}(x, y, z_s, t) \exp\left[i\Delta \phi(x, y, z_s, t)\right] \exp\left[-a_0 L/2\right] \) where \( E_{in}(x, y, z_s, t) \) is the electric field pattern of the incident beam at the sample plane \((z = z_s)\). Taking into account that

\[
I(x, y, z_s, t) = c\varepsilon_0 n_0 \left| E(x, y, z_s, t) \right|^2 \]

the induced phase shift \( \Delta \phi(x, y, z_s, t) \) can also be written as

\[
\Delta \phi(x, y, z_s, t) = \Delta \phi_0^{(3)}(z_s, t) \exp \left( -\frac{2x^2}{w_x^2(z_s)} - \frac{2y^2}{w_y^2(z_s)} \right) + \Delta \phi_0^{(5)}(z_s, t) \exp \left( -\frac{4x^2}{w_x^2(z_s)} - \frac{4y^2}{w_y^2(z_s)} \right)
\]

where \( \Delta \phi_0^{(3)}(z_s, t) = -\kappa_2 I_0(z_s, t) L_{\text{eff}}^{(3)} \) and \( \Delta \phi_0^{(5)}(z_s, t) = -\kappa_2 I_0^2(z_s, t) L_{\text{eff}}^{(5)} \) are the on-axis phase shifts due to the third- and fifth-order nonlinearities respectively. Here \( I_0(z_s, t) \) is the on-axis intensity of the incident beam.

Expanding the phase term \( i\Delta \phi(x, y, z_s, t) \) into a Taylor series and assuming that \( \Delta \phi_0^{(5)}(z_s, t) \ll \Delta \phi_0^{(3)}(z_s, t) \), the electric field at the exit plane of the sample becomes

\[
E_e(x, y, z_s, t) = E_0(t) \left( \frac{w_{0x}w_{0y}}{w_x(z_s)w_y(z_s)} \right)^{1/2} \exp\left(-\frac{a_0 L}{2}\right) \exp[-ikz_s + i\theta(z_s)] \sum_{m=0}^{\infty} \left[ i\Delta \phi_0^{(3)}(z_s, t) \right]^m \frac{m!}{m!} \]

\[
\times \Delta \phi_0^{(5)}(z_s, t) \exp \left[ \frac{(2m+1)x^2}{w_x^2(z_s)} - \frac{(2m+1)y^2}{w_y^2(z_s)} - \frac{ikx^2}{2R_x(z_s)} - \frac{iky^2}{2R_y(z_s)} \right] + \frac{m^m}{m!} \left[ \Delta \phi_0^{(3)}(z_s, t) \right]^{m-1}
\]

Each term in the sum over \( m \) can be associated with two elliptic Gaussian beams with principal semiaxes \( w_{(m1)}^{(m1)} = w_{x,y}(z_s) / \sqrt{2m+1} \) for the beam associated to the first subterm and \( w_{(m2)}^{(m2)} = w_{x,y}(z_s) / \sqrt{2m+3} \) for the beam associated to the second subterm. The radii of curvature for these beams are the same with those of the incident beam, namely \( R_{x,y}(z_s) = R_{x,y}(z_s) \). These relationships, together with equations (2), (3) can be used for determining the location of the waists and the Rayleigh lengths for each individual Gaussian beam, which are

\[
z_{0x,y}^{(m1,2)} = z_s \left( \frac{A_{x,y}^{(m1,2)}}{A_{x,y}^{(m2,2)}} \right)^2 R_{x,y}(z_s) \quad \text{and} \quad z_{0x,y}^{(m1,2)} = z_s \left( \frac{A_{x,y}^{(m1,2)}}{A_{x,y}^{(m2,2)}} \right)^2 R_{x,y}(z_s)
\]

\[
z_{x,y}^{(m1,2)} = z_s \left( \frac{A_{x,y}^{(m1,2)}}{A_{x,y}^{(m2,2)}} \right)^2 R_{x,y}(z_s) \quad \text{and} \quad z_{x,y}^{(m1,2)} = z_s \left( \frac{A_{x,y}^{(m1,2)}}{A_{x,y}^{(m2,2)}} \right)^2 R_{x,y}(z_s)
\]

where \( A_{x,y}^{(m1)} = kw_{x,y}(z_s) / (4m+2) \) and \( A_{x,y}^{(m2)} = kw_{x,y}(z_s) / (4m+6) \). Equations (7) fully define each Gaussian beam, which can be easily propagated to any desired distance \( D \) from the exit plane of the sample according to equations (1) - (4). Finally, their sum gives the total electric field pattern of the laser beam at this distance.
3. Simulations, practical considerations and conclusions

The above results can be used for simulating Z-scan experiments based on either the measurement of the transmittance through an aperture or the direct measurement of the beam dimensions in the far field. However, since most researchers are better acquainted with transmittance measurements and due to the limited space of the paper, in the special case of a circular Gaussian beam only simulations of the transmittance variations are shown (figure 1). On the other hand, when elliptic (and especially astigmatic) beams are considered the direct measurement of the beam dimensions is much more efficient than the measurement of the transmittance through an aperture [3]. Therefore, in this case only simulations of the lengths of the principal semiaxes are depicted (figure 2).

As shown in the above figures the main effect of the higher-order nonlinearities is to modify the distance between the peaks and valleys of the curves. Consequently, it is not easy to decide from a single set of experimental data whether higher order nonlinearities are involved or not. Thus, we suggest the performance of several z – scan experiments for different values of the beam intensity. Based on these data the values of $n_2^{(3)}$ as a function of the intensity can be obtained, using the standard theory for third-order nonlinearity [1-3]. If all these values are constant we conclude that only third- order nonlinearities are involved and no further elaboration is needed. However if above a certain value of the intensity, the $n_2^{(3)}$ values begin to increase or decrease we conclude that the fifthorder nonlinearities are also involved. In this case the theory developed above can be used for calculating the fifth-order nonlinear refractive index $n_2^{(5)}$ in the region where $\left|\Delta\phi_0^{(5)}\right|<<\left|\Delta\phi_0^{(3)}\right|$.

In conclusion we have developed a theoretical infrastructure for analyzing z-scan experiments involving higher-order refractive nonlinearities, and based on simulations we have suggested an experimental procedure for measuring high-order nonlinearities.

4. References

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