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Influence of the photoinduced focal length of a thin nonlinear material in the Z-scan technique

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Abstract: In this paper the response purely refractive of a thin nonlinear material, in the z-scan technique experiment, is modeled as a lens with a focal length that is a function of some integer power of the incident beam radius. We demonstrate that different functional dependences of the photoinduced lens of a thin nonlinear material give typical z-scan curves with special features. The analysis is based on the propagation of Gaussian beams in the approximation of thin lens and small distortion for the nonlinear sample. We obtain that the position of the peak and valley, the transmittance near the focus and the transmittance far from the Rayleigh range depend on the functional dependence of the focal length. Special values of the power reproduce the results obtained for some materials under cw excitation.

OCIS codes: (190.4400) Nonlinear optics, materials; (190.5940) Self-action effects

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1. Introduction

Z-scan technique is a powerful method that has been used to obtain both the sign and magnitude of the complex nonlinear refractive index of some optical materials. The technique is based on the principle that spatial variations of the incident intensity distribution can photoinduce a lens in the nonlinear material which affects the posterior propagation of the beam and intensity changes at far field are obtained. The on axis intensity normalized to that without nonlinear material is called the transmittance. If the transmittance of the nonlinear material is measured as a function of the sample position *z* a characteristic z-scan curve can be obtained. The magnitude and sign of the nonlinearity can be evaluated from the difference between the maximum and minimum transmittance and the shape of the curve, respectively.

Originally, a pulsed Gaussian beam incident to a thin Kerr nonlinear sample was considered to obtain simple analytical formulas relating the z-scan curve obtained from the on axis intensity at the far field. Gaussian decomposition method has been used to analyze the characteristics of the z-scan curves for thin samples with small or large nonlinear phase shifts.

Since the z-scan technique was developed [1, 2], and due to its sensitivity and simplicity, many improvements and modifications have been suggested; between them: eclipsing [3], top hat beams [4], two color [5], reflection [6], etc. A very detailed analysis of the parameters that affect the z-scan measurements was reported by Chapple, *et al*., [7].

Nevertheless all the improvements, modifications and theory developed around the z-scan technique, it exists experimental results, in the thin sample approximation, that are far from the predictions (as example see [8, 9]). This can be due that, in order to apply the z-scan formulas, it is necessary to assume that the material response in a single way to the incident beam, however the response can be the contribution of more than one effect and can not be separated [10,11].

In this paper we study the effect of the focal length of the photoinduced lens in a thin nonlinear material on the z-scan technique. The influence of the nonlinear material is considered as a thin lens with a focal length that depends on a real power *m* of the incident beam radius. Under this assumption, a simple model based on the propagation of a Gaussian beam, in the small phase distortion approximation, through a thin sample is analyzed. Obtaining the normalized transmittance, at the far field, which features depend on the focal length of the photoinduced lens in the nonlinear media. The weak lens limit case is initially analyzed in order to obtain analytic formulas for the peak valley position and transmittance difference as functions of *m*.

2. Model

Considering that at $z=0$ we know the beam waist w_0 of the Gaussian beam used to implement the z-scan technique; the thin nonlinear sample can be modeled as a thin lens of focal length F located at a distance z, further that the photodetector, with a small aperture, is located at a distance L (see Fig. 1), and that the different elements of the optical set up does not change the Gaussian distribution, then we can describe the propagation of the beam using the ABCD law.

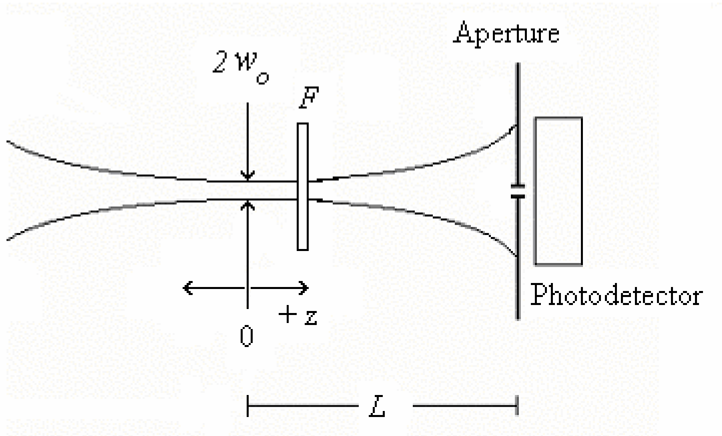

Fig. 1. Z-scan technique scheme.

Assuming only on axis intensity is detected and that the position of the photodetector fulfills the far field approximation, i.e. $L >> z_0$, where z_0 is the Rayleigh distance given by $z_0 = \pi w_0^2 / \lambda$, with λ the wavelength of the beam. Then it is possible to obtain the normalized transmittance of the z-scan experiment as [12]:

$$T = \frac{F^2}{z_0^2 + (F - z)^2} . \tag{1}$$

The above expression is quite general in the sense that no particular form of F has been assumed. In the following we going to obtain the form of F for a Kerr media of thickness d, with refractive index

$$n = n_0 + n_2 I , \tag{2}$$

where n_0 and n_2 are the linear and nonlinear refractive index, respectively. Considering that this sample is illuminated by a Gaussian beam, with intensity

$$I(r,z) = \frac{2P}{\pi w^2(z)} exp\left[-\frac{2r^2}{w^2(z)} \right], \tag{3}$$

where P is the total power and $w(z)$ is the beam radius,

$$w(z) = w_0 \left(1 + (z / z_0)^2 \right)^{1/2} , \tag{4}$$

then this beam, in the parabolic approximation, i.e.

$$I(r,z) \approx \frac{2P}{\pi w^2(z)} \left[1 - \frac{2r^2}{w^2(z)} \right], \tag{5}$$

going to photoinduce a refractive index with a quadratic radial dependence

$$n \approx n_0 + \frac{2n_2 P}{\pi w^2} - \frac{4n_2 P}{\pi w^4} r^2 , \tag{6}$$

where r is the radial coordinate.

This type of refractive index is known as a lenslike and therefore the material has associated an ABCD matrix of the form [13]

$$\begin{bmatrix} A & B \\ C & D \end{bmatrix} = \begin{bmatrix} \cos\gamma d & \dfrac{\sin\gamma d}{n_0'\gamma} \\ -n_0'\gamma\sin\gamma d & \cos\gamma d \end{bmatrix} \tag{7}$$

where

$$\gamma^2 = n_2'/n_0', \quad n_0' = n_0 + \frac{2n_2P}{\pi w^2} \quad \text{and} \quad n_2' = \frac{8n_2P}{\pi w^4}, \tag{8}$$

from the theory of ABCD matrices, the focal length of the system is given by $f = -1/C$. Then considering a thin sample, i.e. when *d* tends to 0, the focal length of the Kerr media is given by

$$F_{Kerr} = \frac{1}{8n_2dP} w^4, \tag{9}$$

where a dependence on the beam radius to the fourth power is obtained.
For a thin thermal media it has been demonstrated that the focal length photoinduced by a Gaussian beam is given by [14]

$$F_{ther} = \frac{\pi\kappa}{P_{abs}(\partial n/\partial T_T)} w^2, \tag{10}$$

where κ is the thermal conductivity, P_{abs} is the absorbed power, $(\partial n/\partial T_T)$ is the change of refractive index with the temperature. In this case the focal length depends on the second power of the beam radius.

From the previous examples we can think that a nonlinear medium, with a refractive nonlinearity, illuminated by a Gaussian beam can be modeled as a lens with a focal length that depends on the beam radius to some integer power, i. e.

$$F = a_m w^m(z), \tag{11}$$

where a_m is a constant with the adequate units, it can have parameters of the material, and *m* is an integer number.

In this paper we going to present an analysis considering individual effects due to different values of *m*, however, some materials can exhibit a nonlinear response than can be probably modeled as the sum of more than one dependence of *w(z)* on *m* [15].

3. Weak lens approximation

We can reduce Eq. (1), if we considered that $F \geq z_0, z$, approximation that we call weak lens, obtaining the following expression:

$$T = 1 + \frac{2z}{F}. \tag{12}$$

Substituting *F,* and *w(z)*, we can rewrite Eq. (12) as;

$$T = 1 + \frac{2z}{F_{0m}\left(1 + (z/z_0)^2\right)^{\frac{m}{2}}}, \tag{13}$$

where $F_{0m} = a_m w_0^m$ is the shortest focal length of the photoinduced lens. From this expression it is possible to calculate the position of the peak and valley of the z-scan curve, giving the following relation

$$\Delta z_{p-v} = \left| z_{peak} - z_{valley} \right| = \frac{2}{\sqrt{m-1}} z_0. \tag{14}$$

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Knowing these positions it is possible to calculate the difference between the transmittance at the peak and valley to obtain:

$$\Delta T_{p-v} = \left| T_{peak} - T_{valley} \right| = \frac{2\Delta z_{p-v}}{F_{0m}} \left[\left(\frac{m-1}{m} \right)^{m} \right]^{1/2} = \frac{2k}{a_m w_0^{m-2}} \left[\frac{(m-1)^{m-1}}{m^m} \right]^{1/2}, \tag{15}$$

where $k = 2\pi / \lambda$.

These last expressions restrict the minimum value of m to be larger than 1 in order to obtain real positions and transmittances, however in Eq. (13) the value $m=1$ produces also a nonconstant transmittance with z that no presents a peak or valley. Note that Δz_{p-v} and ΔT_{p-v} depend in a complicated way on the value of m, besides ΔT_{p-v} is proportional to the inverse of w_0^{m-2}.

In Fig. 2 we plot Eq. (13) for different values of m. The parameters were adjusted to have the same value of ΔT_{p-v} in order to see the main features of the curves for different values of m. As it can see the curves follow the typical shape of a z-scan curve, except for m=1: a prefocal minimum and a post focal maximum, for a positive nonlinearity (the opposite for the negative one), located in a symmetric position with respect to z=0 and similar amplitude with respect to 1. However, the curves present differences in: the peak and valley position, the slope of the linear part (near the waist of the beam) and the decay or growing of the transmittance in the wings (far from the waist of the beam).

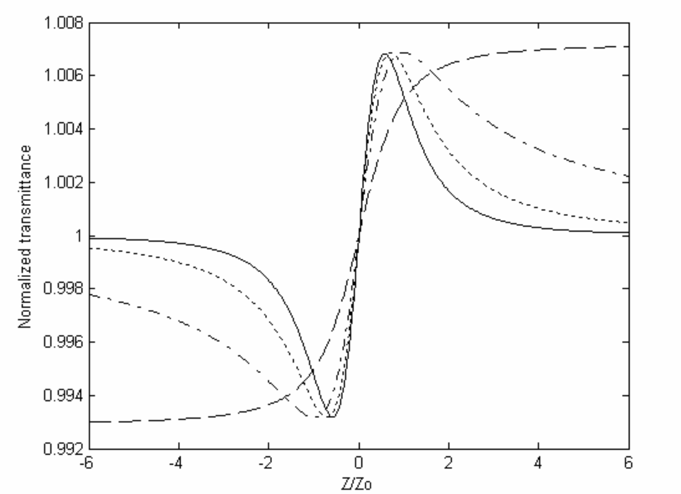

Fig. 2. Z-scan curves for different values of m: 4(solid); 3 (dot); 2 (dashdot) and 1 (dashed). With the following values of the constant: a_4= $1.6\text{x}10^{12}$; a_3=3.8 $\text{x}10^{9}$; a_2= $1\text{x}10^{7}$; a_1=3.8 $\text{x}10^{4}$. w_0 = 20μm and λ=457 nm.

In order to determine the dependence of the normalized transmittance near the focus and far from it we are going to consider the following limit cases:

a) when $|z| << z_0$, in this case the transmittance takes on the following form:

$$T = 1 + \frac{2z}{F_{0m}}, \tag{16}$$

that represents a linear behavior with slope $2 / F_{0m}$, that means that depends inversely on w_0^m.

b) When $|z| >> z_0$, in this case the normalized transmittance takes the following form:

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$$T = 1 + \frac{2z_0^m}{F_{0m} z^{m-1}}, \tag{17}$$

which means that the wings of the curve present an inverse dependence on z^{m-1}. Then depending on the value of *m*, the normalized transmittance reaches the peak or valley in a faster or slower way.

From the above analysis is clear that the value of *m* will determine the main features of the z-curve because it define the separation between the peak and valley, the dependence of ΔT_{p-v} with the beam waist and the dependence of the normalized transmittance in the wings with z. Next we present in detail some special values of *m*. Note that any real value can be used, however we are going to restrict to only integers.

3.1 Special case m=4.

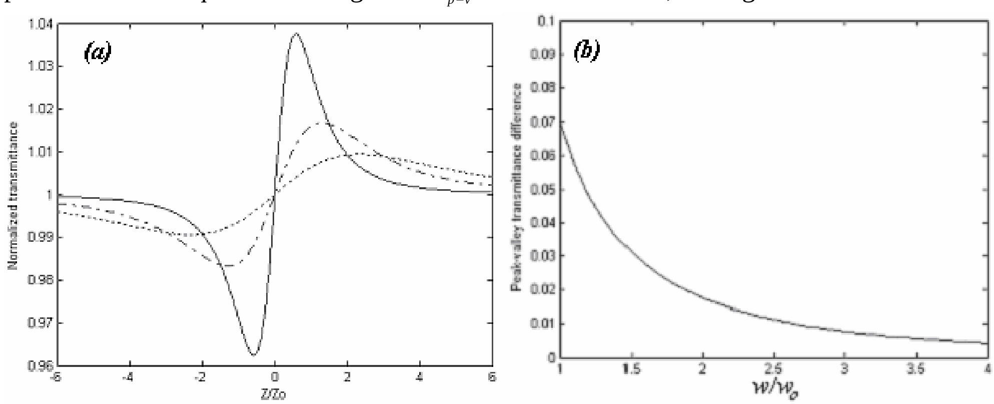
For this value of *m* we obtain the following relations:

$$\Delta z_{p-v} = \frac{2}{\sqrt{3}} z_0 \quad , \ \Delta T_{p-v} = \frac{3\sqrt{3}}{8} \frac{k}{a_4 w_0^2},$$

$$T = 1 + \frac{2z}{F_{04}} \text{ for } |z| << z_0, \text{ and } T = 1 + \frac{2z_0^4}{F_{04} z^3} \text{ for } |z| >> z_0,$$

where $F_{04} = a_4 w_0^4$.

The value of Δz_{p-v} agrees very well with that reported in the small distortion approximation of a sample with a Kerr nonlinearity [16, 17]. The transmittance in the wings of the curve present a dependence inverse with z^3. It is important to note that $\Delta T_{p-v} \propto w_0^{-2}$, for this value of *m*. Then changing the beam waist and using the same sample and incident power an inverse quadratic change of ΔT_{p-v} must be obtained, see Fig. 3.



(a) *(b)*

Fig. 3. (a). Z-scan curves for m=4 and different beam waists: w_0 (solid); $1.5w_0$ (dashdot) and $2w_0$ (dot) $a_4 = 3.2\text{x}10^{11}$; $w_0 = 20\ \mu$m and λ=457 nm. b) ΔT_{p-v} as function of the beam waist.

3.2 Special case m=2

In this case we obtain the following relations:

$$\Delta z_{p-v} = 2z_0, \ \Delta T_{p-v} = \frac{k}{a_2},$$

$$T = 1 + \frac{2z}{F_{02}} \text{ for } |z| << z_0, \text{ and } T = 1 + \frac{2z_0^2}{F_{02} z}, \text{ for } |z| >> z_0,$$

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where $F_{02} = a_2 w_0^2$.

The above results were reported in Ref. [12]. ΔZ_{p-v} increase with respect to the case of *m=4*, in fact this value coincide with references [18,19]. The transmittance in the wings present a dependence inverse with *z*. $\Delta T_{p-v} = k / a_2$ does not depend on the beam waist, then the z-scan curves with different lenses, keeping all the other parameters constant, must have the same transmittance difference, see Fig. 4. This fact represent a significant difference with respect to the case *m*=4.

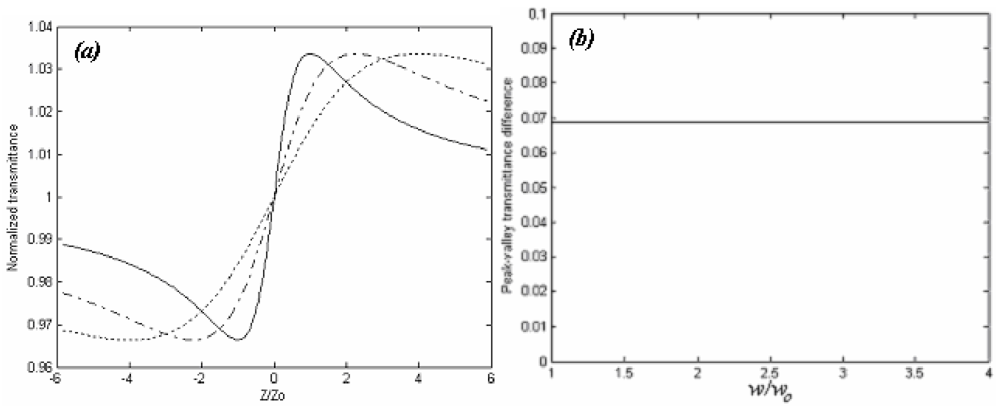

Fig. 4. Z-scan curves for m=2 and different beam waists: w_0 (solid); $1.5w_0$ (dashdot); $2w_0$ (dot). $a_2 = 2x10^6$; $w_0 = 20\mu m$. ΔT_{p-v} is independent of the beam waist. b) ΔT_{p-v} as function of the beam waist.

3.3 Special case m=3

In this case

$$\Delta z_{p-v} = \sqrt{2} z_0 , \; \Delta T_{p-v} = \frac{4}{3\sqrt{3}} \frac{k}{a_3 w_0}$$

$$T = 1 + \frac{2z}{F_{03}} \text{ for } |z| << z_0 \text{, and } T = 1 + \frac{2z_0^3}{F_{03} z^2} \text{, for } |z| >> z_0 ,$$

where $F_{03} = a_3 w_0^3$.

For this value of *m*, Δz_{p-v} is smaller than that obtained for *m*=2 and larger than that for *m*=4. The transmittance in the wings follows a dependence inverse with z^2. The dependence of ΔT_{p-v} is inverse linear with w_0, see Fig. 5.

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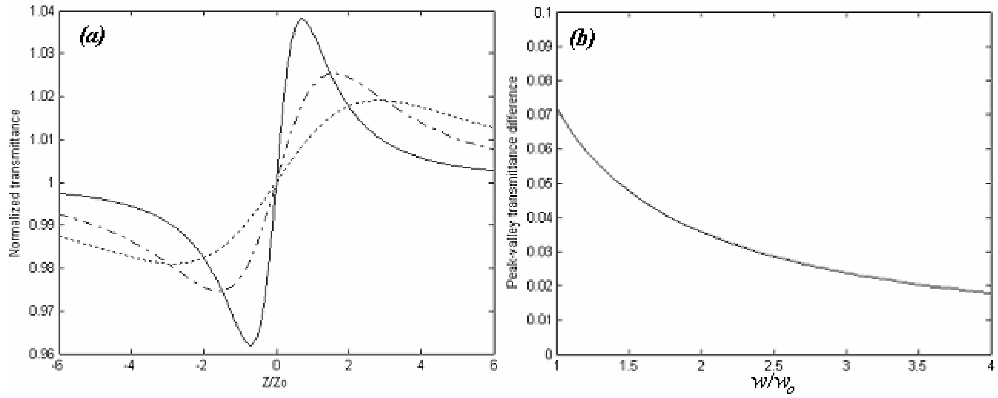

Fig. 5. Z-scan curves for m=3 and different beam waists: w_0 (solid); $1.5w_0$ (dashdot); $2w_0$ (dot). $a_3 = 7.4 \times 10^8$; $w_0 = 20\mu m$. b) ΔT_{p-v} as function of the beam waist.

3.4 Special case m=1

In this case the curve does not exhibit a peak and valley, however the normalized transmittance is not constant, then it is possible to obtain a normalized transmittance difference that have the following dependence with*:*

$$\Delta T = 2\frac{w_0 k}{a_1},$$

that represents a linear dependence with w_0. In Fig. 6 we plot the normalized transmittance for different beam waists. Note that the curves continue being very symmetric.

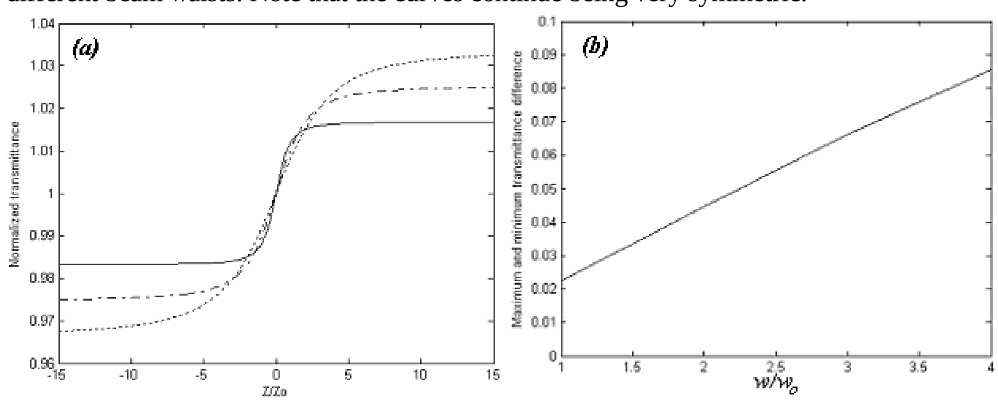

Fig. 6. Z-scan curves for m=1 and different beam waist: w_0 (solid); $1.5w_0$ (dashdot) and $2w_0$ (dash). b) ΔT_{p-v} as function of the beam waist.

4. Complete formula

It is necessary to use Eq. (1) when the minimum photoinduced focal length F_{0m} is of the same order or smaller than z_0. In this case is not possible to obtain formulas for the peak-valley position and transmittance difference, as in the weak approximation. In the following analysis we restrict the maximum value of the normalized transmittance to 5 in all the curves, greater values means phase differences on axis greater than 2π, then light from different radial distances of the beam could interfere losing the beam its Gaussian distribution.

The curves obtained for different values of *m* present a sharp peak and a broad valley. The peak moves to the right in the case of a positive photoinduced lens and the opposite for a

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negative one. The valley moves to the position $z=0$. As consequence, ΔZ_{p-v} grew as F_{0m} decreased. In general this case was characterized by asymmetric curves.

In order to demonstrate that the parameter m continues determining some features of the z-scan curves, we present typical results obtained for different values of m. For m = 4, and different values of the ratio F_{0m}/z_0, see Fig. 7, we can see that the peak is very sharp compared with the valley. The normalized transmittance in the valley, in some cases, almost reached the value of zero and it was located at z=0. The normalized transmittances in the wings almost reach the value of one. Note that the same behavior for the z-scan curves was reported in [20] for a thin film of amorphous As_2S_3 considering large phase shifts. They also reported a formula (Eq. 11 in Ref. [20]) for the normalized transmittance in terms of the nonlinear phase shift $\Delta\Phi_0$; that can be reproduced after some algebraic manipulation of our Eq. 1 to give

$$T = \frac{1}{1-\frac{4x}{(1+x^2)^2}\left(\frac{z_0}{2F_{0m}}\right)+\frac{4}{(1+x^2)^3}\left(\frac{z_0}{2F_{0m}}\right)^2},$$

where $x = z/z_o$ and the relation between $\Delta\Phi_0$ and our parameters is;

$$\Delta\Phi_0 = \frac{z_0}{2F_{04}}.$$

By comparison, we can generalize and define the nonlinear phase shift on axis in our model as:

$$\Delta\Phi_{0m} = z_0/2F_{0m}.$$

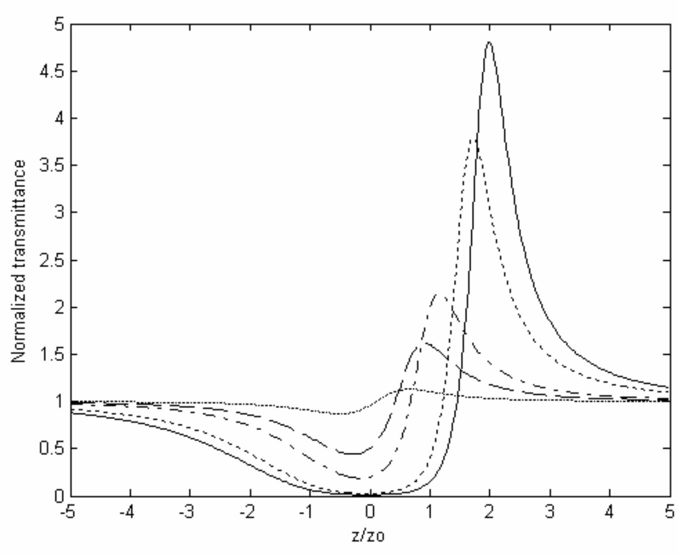

Fig. 7. Z-scan curves for m=4 and different values of F_0/z_0: 5 (point), 1 (dash), 0.5 (dashdot), 0.16 (minus sign) and 0.11 (solid).

When the waist of the incident beam was changed the peak-valley transmittance difference was reduced as the waist was increased but not at the ratio obtained for the weak lens approximation. However the difference is clear, Fig. 8.

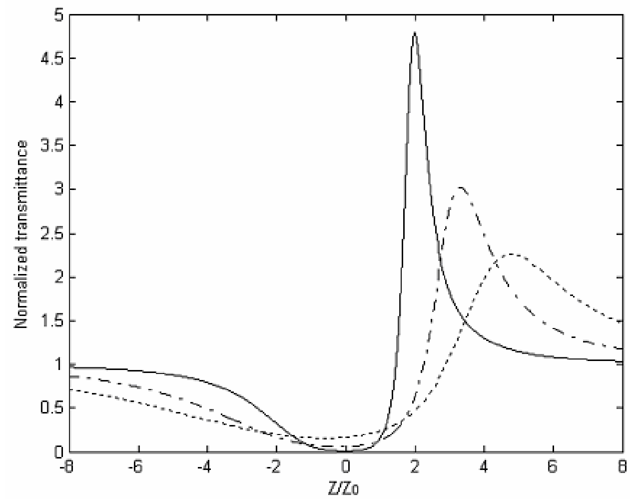

Fig. 8. Z-scan curves for m=4, a_4= 1.8x10^9, and different beam waist: w_0 (solid);1.5w_0 (dashdot) and 2w_0 (dash).

The same analysis but now with *m*=2 gave z-scan curves with different features. The peak was not as sharp as in the case of *m=4* and the valley never reach the value of zero. The normalized transmittance in the wings was different for each curve: positive values of *z* gave greater differences than the negative ones, Fig. 9. When the waist of the beam was changed the obtained peak-valley transmittance difference was practically the same. This result was the same than that obtained in the weak lens approximation, then this characteristic is maintained for this value of *m*, Fig. 10.

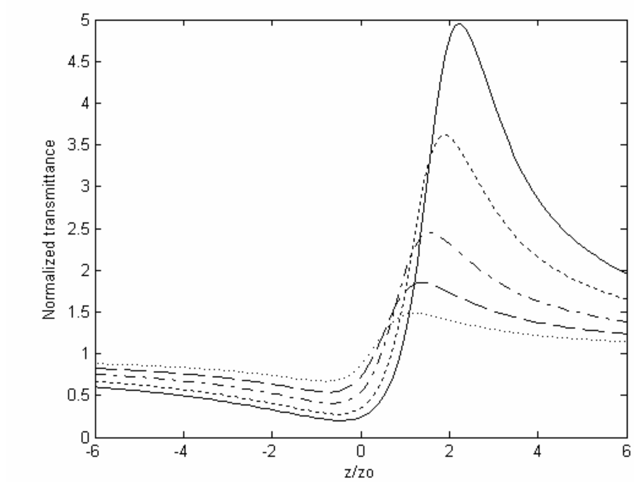

Fig. 9. Z-scan curves for m=2, and different values of F_0/z_0: 2.5 (dot), 1.6 (dash), 1 (dashdot), 0.72 (minus-sign), 0.56 (solid).

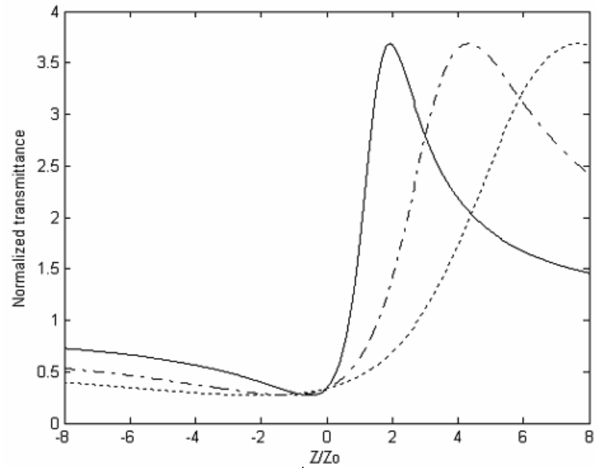

Fig. 10. Z-scan curves for m=2, a_2 = 4.9x10^4 and different beam waists: w_0 (solid); 1.5w_0 (dashdot) and 2w_0 (dash).

For *m=1*, the curves obtained for different values of the ratio F_0/z_0 are shown in Fig. 11. They are asymmetric and small changes in the ratio produce large amplitude curves.

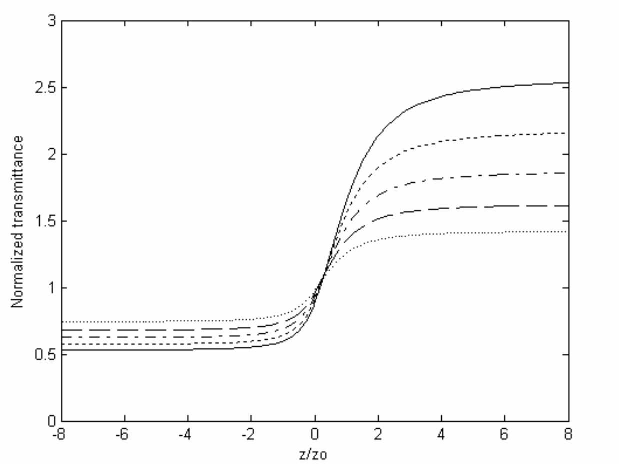

Fig. 11. Z-scan curves for m=1 and different values of F_{01} / z_0 : 2.6 (solid), 3.1 (minus sign), 3.7 (dash-dot), 4.6 (dash) and 6.2 (point).

ΔT_{p-v} is the parameter that has been used to evaluate the magnitude of the nonlinearity of the tested sample. We obtain that different values of *m* gave the same ΔT_{p-v} when $F_{0m} / z_0 > 10$, differences are obtained when F_{0m} / z_0 is smaller, see Fig. 12.

ΔZ_{p-v} as function of F_{0m} / z_0 presents a clear dependence with *m*, see Fig. 13. Values of $F_{0m} / z_0 > 6$ gave a magnitude of ΔZ_{p-v} that follows the relation obtained by Eq. 12, weak lens approximation, for each *m*. While values of $F_{0n} / z_0 <6$ gave larger differences, however it is possible to associate a unique value of *m* depending on the F_{0m} / z_0 ratio**.**

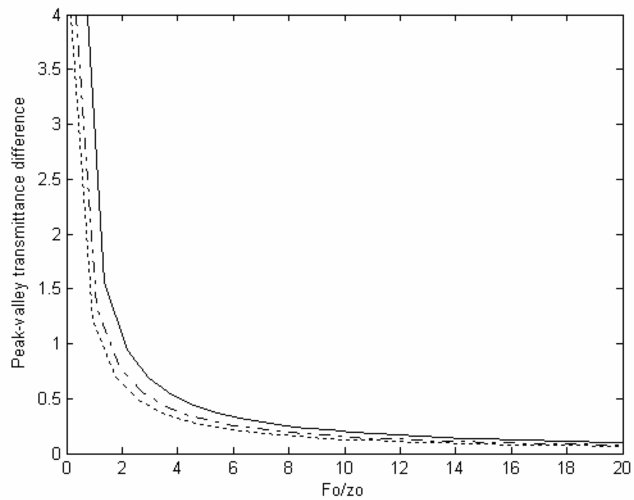

Fig. 12. ΔT_{p-v} as function of F_0 / z_0 and different values of m: 2(solid), 3(dashdot) and 4(dot).

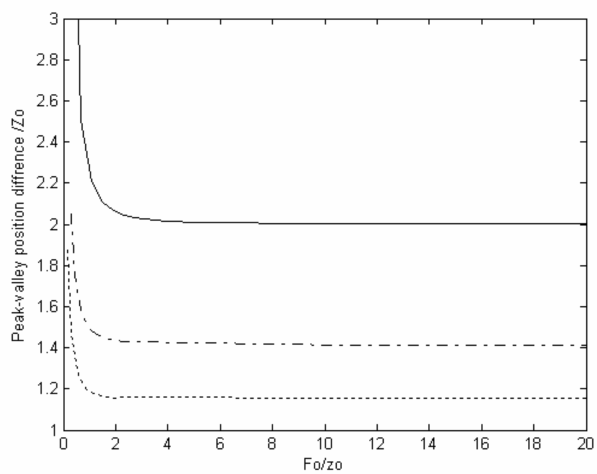

Fig. 13. ΔZ_{p-v} as function of F_0 / z_0 for different values of m: 2 (solid), 3 (dashdot) and 4 (dot).

5. Conclusion

In conclusion we have analyzed the influence of the focal length of the photoinduced lens in a nonlinear material in the Z-scan technique. The focal length of the photoinduced lens was considered as dependent on the incident beam radius *w(z)* to some integer power *m*. Gaussian beam propagation and thin lens approximation were used to obtain an expression for the on axis far field normalized transmittance. The obtained z-scan curves present different features according to the value of *m*. Approximation to weak lens allowed to obtain analytic formulas for the peak-valley position and transmittance difference. Showing that, the peak-valley position difference is strongly dependent on the value of *m*. Another parameter that is clearly affected by *m* is the peak-valley transmittance difference for different beam radius used. Then it is not necessary to suppose what dependence, on the photoinduced lens, will present the sample to characterize, this can be determined if the waist of the beam is known or analyzing the change in transmittance difference for different focusing lenses.

The model presented here can be completed to include nonlinear absorption or to describe the transmitted beam using Gaussian beam decomposition or to include aberration of the photoinduced lens in order to describe more real samples and experimental conditions. However this can be used as a first approximation to explain experimental results were the type of photoinduced lens is not known.

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