Theory of Thin Sample z-scan of a New Class of Nonlinear Materials

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Abstract - We report the theory of thin-sample z-scan for materials, viz. diffusion-dominated photorefractives, having a nonlinearly induced phase that may be proportional to the spatial derivative of the intensity profile. The on-axis far-field intensity is approximately an even function of the scan distance on different positive and negative values for phase shift $\Delta\Phi_o$. In case of positive phase shift, the z-scan graph shows a minimum and two maxima, while for the negative value, only one minimum is observed. The fact is that far-field beam profiles display beam distortion and shift of the peak as compared with Kerr-type or photovoltaic nonlinearities.

Keywords: z-scan, thin-sample, nonlinearity, diffusion-dominated photorefractive, photovoltaic

1. Introduction

The z-scan method is a highly sensitive, simple, and versatile technique for measuring the sign and the magnitude of light-induced variations of the refractive index of nonlinear optical materials [1]. Much research has been done in the area of thin and thick sample z-scan of Kerr-type materials [2-3], with cascaded nonlinearities [4], thermal nonlinearities [5], photovoltaic nonlinearities and inhomogeneous and anisotropic nonlinearities [6-8]. In general, a light beam focused into a nonlinear material undergoes both phase and amplitude changes during propagation giving rise to a revised profile in the far-field. By measuring far-field on-axis intensity or beam width as a function of scan length around the rear focal plane of a lens, the sign and magnitude of the effective nonlinear refractive index can be calculated. In some cases, measurement of beam profile and/or beam ellipticity can provide valuable information regarding the nonlinearity, as seen for instance in the case of inhomogeneousely induced nonlinearities, due to photovoltaic effect in photorefractives. Photorefractive materials have been widely studied [6-9] and used in applications ranging from holographic memories, beam coupling, phase conjugation, etc. There are two kinds of induced nonlinearities in a photorefractive material. One is the diffusion-dominated type $\Delta n \;\alpha\; \nabla I, e.g., BaTiO_3$. These materials have the characteristic of the nonlinearly induced phase shift that may depend upon the spatial derivative of optical intensity.

The other type is $\Delta n \;\alpha\; I, e.g., LiNbO_3$, the photovoltaic type. In previous papers [10-11], we have discussed z-scan and p-scan for photovoltaic materials. It was shown that the z-scan graph has the same approximate shape as that for materials with Kerr-type nonlinearity and the effective nonlinear refractive index coefficient was determined.

In this paper, we have developed the theoretical z-scan of diffusion dominated materials where the induced nonlinearity is of the form $\Delta n \propto \nabla I$, which depends upon the spatial derivative of optical intensity. We show: (a) first the z-scan and the beam profile distribution in the far field for Kerr-type materials for comparison; (b) the expression for the far field beam profiles during z-scan of materials where $\Delta n \propto \nabla I$; (c) the z-scan plots along with the far field intensity profiles in this case; (d) comparison with a simple physical picture; (e) differences between positive and negative "derivative" nonlinearities.

2. Theoretical Model

Assuming a TEM_{00} radially symmetric Gaussian beam of beam waist radius w_o traveling in the + z direction, the electric field E can be written as:

$$E(r,z) = \mathrm{Re}\left\{E_o \frac{w_o}{w(z)} \exp\left[-r^2\left(\frac{1}{w^2(z)} + \frac{ik}{2R(z)}\right)\right] \exp[-i\Phi(z,t)]\right\}$$
$$= \mathrm{Re}\{E_e(r,z)\exp[-i\Phi(z,t)\} \quad (1)$$

where E_e denotes the complex envelope of E and is a slowly varying function of z, $w^2(z) = w_o^2[1 + z^2 / z_o^2]$ is the

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beam radius, $R(z)=z[1+z_o^2/z^2]$ is the radius of curvature of the wave front at z, $z_o=kw_o^2/2$ is the Rayleigh length of the beam, $k=2\pi/\lambda_0$ is the propagation constant in free space and λ_0 is the laser wavelength. E_o represents the electric field at the focus defined by $w(z)=w_0$ and on the optical axis. The $\exp[-i\Phi(z,t)]$ term contains all the radially uniform phase variations. For a Kerr-type optical nonlinearity, the refractive index of the nonlinear material with a Gaussian beam can be written as:

$$n=n_0+n_2|E_e|^2 \tag{2}$$

where n_0 is the linear refractive index. A more general representation intensity-dependent refractive index can be expressed in the form $n=n_0+\Delta n(I)$ where I is the optical intensity or irradiance. In order to analyze the z-scan data, we need to calculate the electric field E at the observation plane for any scan distance z of the sample. For analyzing this electric field distribution, the nonlinear paraxial wave equation which is similar to the nonlinear Schrodinger equation for a Kerr-type nonlinearity must be solved within the sample. If the sample length is much smaller than the Rayleigh range, and if the phase changes in the field caused by the nonlinear interaction are not transformed into amplitude changes within the sample, then the sample is considered to be thin (external self-action) [2, 12].

By applying the slowly varying envelope approximation and the thin-sample approximation [12, 14], the induced phase of the electric field during propagation through the nonlinear sample can be written as:

$$\frac{d\Delta\Phi}{dz'}=+\Delta n(I)k \tag{3}$$

where z' is the propagation distance within the sample. Here z' should not be confused with sample position z. In case of thin sample and negligible nonlinear absorption, Eq. (3) is solved to give the phase shift $\Delta\Phi$. For a thin sample of thickness L, along which I is almost constant, the phase shift will be $\Delta\Phi(L)=+k\Delta n(I)L$. Since the intensity distribution of the fundamental Gaussian beam is

$$I(r,z)=|E_e(r,z)|^2=E_o^2\frac{w_o^2}{w^2(z)}\exp\left[\frac{-2r^2}{w^2(z)}\right] \tag{4}$$

we can obtain the total phase shift $\Delta\Phi$ by integrating Eq.(3), using Eq.(4),

$$\Delta\Phi(r,z)=\Delta\Phi_o(z)\exp\left[\frac{-2r^2}{w^2(z)}\right] \tag{5}$$

with

$$\Delta\Phi_o(z)=\frac{\Delta\Phi_o}{1+z^2/z_o^2} \tag{6}$$

where the phase factor, $\Delta\Phi_o=k\Delta n_o L$, is related to the optically induced on-axis phase shift at focus and Δn_o is on-axis nonlinear index change at the focus. The complex electric field with the nonlinear phase shift term at the focus ($z=0$) can be written as:

$$E(r,0)=E(0,0)\exp\left[\frac{-r^2}{w_o^2}-i\Delta\Phi(r,0)\right] \tag{7}$$

Expanding the Gaussian radially dependent portion of this complex phase shift results in the series [1]

$$E(r,0)=E(0,0)\sum_{m=0}^{\infty}\frac{[-i\Delta\Phi_o(0)]^m}{m!}\exp\left\{\frac{-r^2}{w_m^2(0)}\right\} \tag{8}$$

where the radius of each of the individual Gaussian term is given by $w_m^2(0)=w_o^2/(2m+1)$.

The optical field immediately behind the thin nonlinear sample a distance z from the rear focal plane of the external lens is represented by:

$$E(r,z)=E_o\frac{w_o}{w(z)}\exp\left(\frac{-ikr^2}{2r(z)}\right)\sum_{m=0}^{\infty}\frac{[-i\Delta\Phi_o(0)]^m}{m!}\exp\left\{\frac{-r^2}{w_m^2(z)}\right\} \tag{9}$$

where $w_m^2(z)=w^2/(2m+1)$ and all z-dependent values are measured with respect to the rear focal plane of the external lens; $w^2(z)$, $R(z)$ are respectively the width and radius of curvature of the radially symmetric Gaussian beam for arbitrary scan distance z, and $\Delta\Phi_o(z)=\Delta\Phi_o/(1+(z/z_R)^2)^{1/2}$, z_R denotes the Rayleigh length corresponding to w_o. The far-field optical field at detector plane for any sample position z can be evaluated by Huygens-Fresnel propagation integral [12].

Next, consider the nonlinear effect of the diffusion dominated photorefractive materials described in the introduction. This material has the characteristic of the nonlinearily induced phase shift that may depend upon the spatial derivative of optical intensity (thin-sample with "derivative" nonlinearity). The phase of the optical field immediately behind the sample is, in this case, proportional to the spatial derivative of the intensity profile. Thus,

$$\Delta\Phi(r,z)=\Delta\Phi_o(z)\frac{\partial}{\partial r}\exp\left[\frac{-2r^2}{w^2(z)}\right] \quad (10)$$

At the focus ($z = 0$), the complex electric field can also be written as:

$$E(r,0)=E(0,0)\exp\left[\frac{-r^2}{w_o^2}-i\frac{\partial}{\partial r}\Delta\Phi(r,0)\right] \quad (11)$$

As before, expanding the Gaussian components of these complex phase shifts results in the series

$$E(r,0)=E(0,0)\sum_{m=0}^{\infty}\frac{[-i\cdot b(r)]^m}{m!}\exp\left\{\frac{-r^2}{w^2m(0)}\right\} \quad (12)$$

where $b(r) = -4r / w^2(0)$ is a function of radial variation. The optical field immediately behind the thin nonlinear sample placed at distance z from the focus can be written as:

$$E(r,z)=E_o\frac{w_o}{w(z)}\exp\left(\frac{-ikr^2}{2R(z)}\right)\sum_{m=0}^{\infty}\frac{[-i\Delta\Phi_o(z)]^m[-4r/w^2(z)]^m}{m!} * \exp\{-(2m+1)r^2/w^2(z)\} \quad (13)$$

where all z-dependent values are also measured with respect to the rear focal plane of the external lens; $w^2(z)$, $R(z)$ are respectively the width and radius of curvature of the radially symmetric Gaussian beam for arbitrary scan distance z, z_R denotes the Rayleigh length corresponding to w_o, and also phase factor $\Delta\Phi_o(z)=\Delta\Phi_o/(1+(z/z_R)^2)^{1/2}$.

To analyze the intensity profile for the diffusion dominated photorefractive sample on the observation plane, we also use the Huygens-Fresnel propagation integral [12]. The far-field optical field pattern for any sample position z is

$$E_{ff}(k_r,z)=\int_0^{\infty} rE(r,z)J_0(k_r r)dr \quad (14)$$

where k_r has the connotation of a spatial transverse variable according to $k_r = k_0 r / z_{ff}$, where z_{ff} denotes the distance to the far-field after the sample. Terms in the resulting series can be evaluated by use of [13]

$$\int_0^{\infty} x^u \exp(-\alpha x^2)J_v(\beta x)dx = \frac{\beta^v\Gamma((u+v+1)/2)}{2^{v+1}\alpha^{(u+v+1)/2}\Gamma(v+1)}F_1^1\left\{\frac{(u+v+1)}{2};v+1;\frac{-\beta^2}{4\alpha}\right\} \quad (15)$$

where Γ and F_1^1 are the gamma function and confluent hypergeometric function respectively. To calculate the far-field pattern of the beam at the observation plane we used the above relation. Using the properties of a Gaussian beam [14], the normalized intensity at any point in the observation plane $I(k_r, z)$ is given by:

$$I(k_r,z,\Delta\Phi_o)=\frac{|E(k_r,z,\Delta\Phi_o(z))|^2}{|E(k_r=0,z,\Delta\Phi_o(z=0))|^2} \quad (16)$$

with far-field conditions, which propagation distance in free space from the sample to the aperature plane is much larger than Rayleigh length of the beam z_o. In the following section, we plot z-scan pictures and beam profiles in the far-field for Kerr-type samples (or, photovoltaic dominated photorefractive samples) and also for the derivative nonlinearity of diffusion dominated photorefractive samples.

3. Numerical Results

We have estimated the on-axis beam intensity variation at the origin of observation plane according to the displacement of sample around the focal point of the lens (z-scan). The procedure involves scanning the sample around the rear focal plane of an external lens and observing the far-field pattern. The variation of on-axis transmittance with scan length gives magnitude and sign of n_2. Beam profiles in far-field for a lens with focal length 5 cm, λ_o=514 nm, initial beam intensity diameter 0.77 mm and $\Delta\Phi_o$ (peak on-axis phase shift) = - 0.05 are computed for various scan lengths.

Fig. 1 shows a plot of I (r,z) as a function of z and the radial variation. This 3-D Fig. can be used in a z-scan transmittance measurement to determine the magnitude and sign of n_2. k_r represents a far-field parameter and is equal to $k_r = k_0 r / d$, where d is the distance from the sample to the observation plane in the far field. $k_r = 0$ signifies the on-axis position or r = 0. A typical z-scan behavior for nonlinearity around r = 0 (see Fig. 2 (a)) can be seen with a complimentary behavior for large r (see Fig. 2 (b)), due to energy conservation.

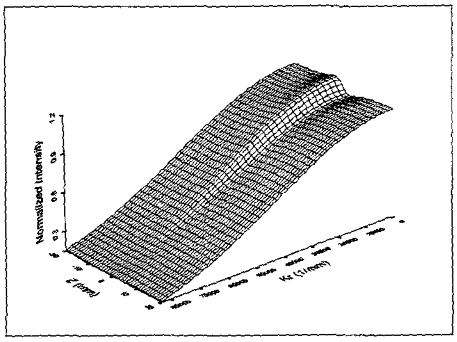

Fig. 1 Calculation of normalized intensity Z -scan curves for Kerr-type samples with $\Delta\Phi_o = -0.05$, f = 0.05 m, λ_o = 514nm, initial beam intensity diameter 0.77mm.

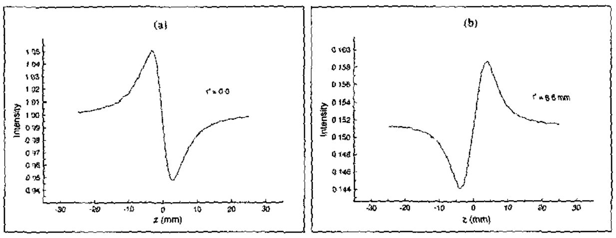

Fig. 2 Calculated Z -scan intensity curves as Kerr-type sample for on-axis variation (a) $r' = 0$, and (b) radial variation $r' = 6.8\,mm$.

For the optical nonlinearity, for instance, in a diffusion dominated photorefractive material, the phase term of the light distribution of the sample needs to be spatially differentiated with respect to the radial coordinate space (thin-sample Z -scan with "derivative" nonlinearity). We have

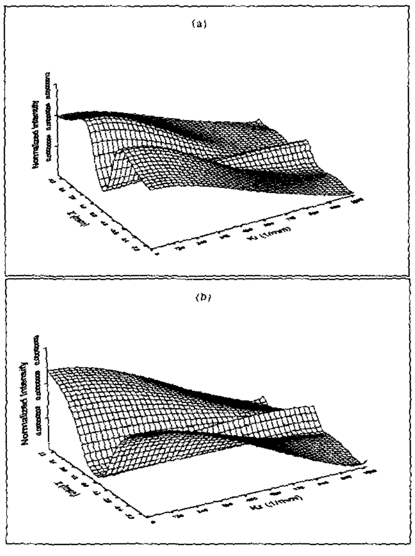

Fig. 3 Beam profiles in far-field for the diffusion dominated photorefractive sample (a) $\Delta\Phi_o = +0.05$ and (b) $\Delta\Phi_o = -0.05$ with f = 0.05 m, λ_o = 514nm, the laser beam intensity diameter 0.77mm.

considered observation-plane characteristics of an initial Gaussian laser beam that passes through a thin sample of such a material, as well as the far-field intensity-distribution in non-Gaussian, as shown Fig. 3 and Fig. 4.

Fig. 3 (a) and 3 (b) show plots of far-field intensity as a function of scan distance z and radial variation $[k_r(1/mm)]$ for thin-sample with positive and negative derivative nonlinearity. These 3-D Fig.s can be used to find the far-field on-axis intensity or the beam profile as a function of scan length around the rear focal plane of the external lens. The on-axis far-field intensity patterns are approximately an even function of the scan distance, see Fig. 3, as well as Fig. 4 drawn for different positive and negative values for $\Delta\Phi_o$.

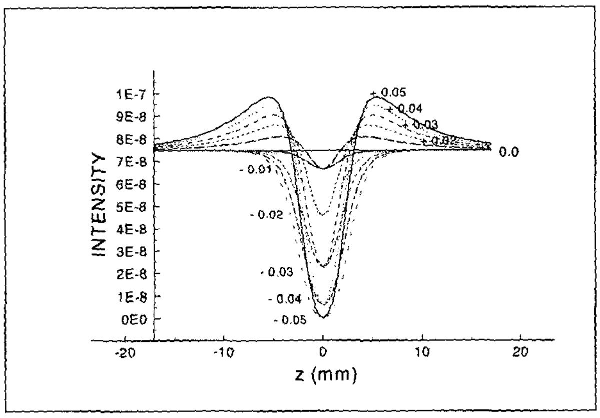

Fig. 4 Calculated Z -scan intensity for diffusion dominated photorefractive sample for variable value $\Delta\Phi_o$ (peak on-axis phase shift).

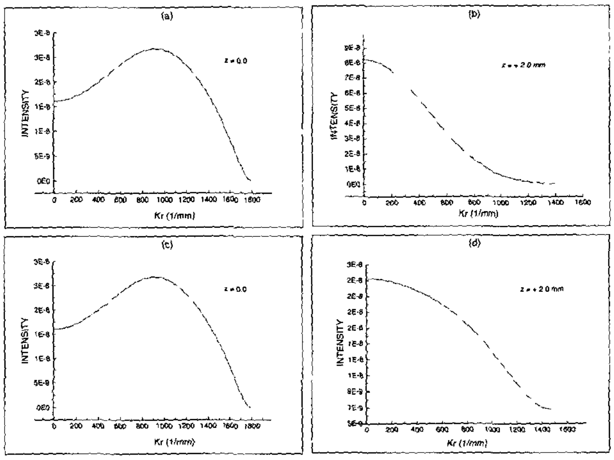

Fig. 5 Calculated Z - scan radial variation intensity curves as "derivative" nonlinear samples with $\Delta\Phi_o = +0.05$ in (a) and (b), $\Delta\Phi_o = -0.05$ in (c) and (d).

In Fig. 4, we plot the on-axis variation of the intensity as a function of scan length for various positive and negative $\Delta\Phi_o$. Note that for positive $\Delta\Phi_o$, the z - scan graph shows one minimum and two maxima, while for negative $\Delta\Phi_o$, only one minimum is observed. The fact that the z-scan is approximately an even function for $\Delta\Phi \propto \nabla I$ (as seen from Fig. 4) can be analytically argued as well (refer to Appendix).

Fig. 5 also shows the calculated radial variation in the far-field of the intensity for a "derivative" nonlinear sample. Note that at $z = 0$, the beam profile has a local minimum on axis for both positive and negative $\Delta\Phi_o$, while for sufficiently large $|z|$, the profile monotonically decreases from the on-axis value.

4. Conclusion

In conclusion, we have estimated the far-field intensity distribution on optical axis as well as beam profile of both z-scan of Kerr-type materials and thin-sample z-scan of materials with derivative nonlinearities. The z-scan picture is symmetric with respect to the scan length z. The shape of the z-scan graphs can be reconciled with a simple physical picture. The sign of the nonlinearity can be determined from the shape of the z-scan graph. The peak to valley intensity ratio depends on the magnitude of the nonlinearity. Extension to anisotropic induced derivative nonlinearities will be pursued in the future. Results should be useful in characterizing the nonlinearities of diffusion dominated photorefractive materials. It may be possible to postulate whether the z-scan graph will be symmetric or nonsymmetric by examining the nature of the induced nonlinearity.

Appendix

Let G' denote a Gaussian function of r, the radial coordinate. The complex field immediately behind the thin sample can be written as the product of a Gaussian G' and a complex exponential $\exp\pm(j\alpha r^2)$ whose argument denotes the quadratic phase due to propagation. Note that the $\pm$ sign corresponds to the phase before and after the geometrical focus of the external lens (i.e., for $z<0$ and $z>0$), respectively. The induced phase is proportional to the gradient of the intensity. The far-field on-axis optical fields for positions of the sample at $z<0$ and $z>0$ can be written in terms of the integrals $I_{1,2}$ respectively as:

$$I_1 = \int_0^\infty G' e^{-j(rG' - \alpha r^2)} r dr \qquad \text{(A1)}$$

$$I_2 = \int_0^\infty G' e^{-j(rG' + \alpha r^2)} r dr \qquad \text{(A2)}$$

In (A2), we put $r' = -r$, and noting that G' is an even function of r, then

$$I_2 = \int_0^{-\infty} G' e^{-j(-r'G' + \alpha r'^2)} r' dr' = \int_0^{-\infty} G' e^{+j(rG' - \alpha r^2)} r dr$$

$$= \int_0^{-\infty} G' e^{-j(rG' - \alpha r^2)} r dr$$

thus

$$|I_1|^2 = \left[\int_0^\infty G' e^{-j(rG' - \alpha r^2)} r dr\right]\left[\int_0^\infty G' e^{+j(sG' - \alpha s^2)} s ds\right]$$

$$|I_2|^2 = \left[\int_{-\infty}^0 G' e^{+j(sG' - \alpha s^2)} s ds\right]\left[\int_{-\infty}^0 G' e^{-j(rG' - \alpha r^2)} r dr\right]$$

Using $I_1 - I_2 = 0$

$$\therefore \int_0^\infty G' e^{-j(rG' - \alpha r^2)} r dr + \int_{-\infty}^0 G' e^{+j(rG' - \alpha r^2)} r dr = 0$$

Since G' is an even function of r,

$$\int_0^\infty G\, e^{-jrG} r dr = 0$$

thus

$$\int_0^\infty G' (\cos rG - j \sin rG) r dr = 2\int_0^\infty G' (\sin rG) r dr = 0$$

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