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Effect of coupling between linear absorption and nonlinear absorption on Z-scan measurements*

Zang Wei-Ping(臧维平), Tian Jian-Guo(田建国)†, Liu Zhi-Bo((刘智波),
Zhou Wen-Yuan(周文远), Song Feng(宋 峰), and Zhang Chun-Ping(张春平)

Photonics Centre, College of Physics, Nankai University, Tianjin 300071, *China*

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The contribution of multiphoton absorption to open-aperture Z-scan measurements is analysed by coordinate transformation. The approximate closed-form solution is obtained, which is suitable for application in higher-optical power regime. As an example, the coupling between linear absorption and two-photon absorption is discussed in detail.

Keywords: Z-scan, coordinate transformation, nonlinear optics, and multiphoton absorption
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1. Introduction

The Z-scan technique has been utilized extensively for measurements of nonlinear index and nonlinear absorption because of its simplicity and accuracy.[1,2] The beam propagation in a thin nonlinear medium has already been analysed and modelled extensively. But the analysis of Gaussian beam propagation in a thick nonlinear medium is usually more complicated. Some analytical approaches have been applied,[3−8] most of them involve the aberrationless approximation, and the results are only correct to the first order in irradiance. Zang analysed the Z-scan within strongly nonlinear regime by a novel variational method,[9] but the numerical calculation just involved a nonlinear medium only with nonlinear absorption. Hermann gave an approximate solution for the strongly nonlinear absorption regime,[10,11] but the algebra was rather tedious and the formula exhibiting a saturation effect was obtained through hypothesis.

In this paper, an approximate closed-form solution to open-aperture Z-scan measurements with multiphoton absorption is obtained by coordinate transformation, which is suitable for application in higher-optical power regime. The effect of the coupling between linear absorption and nonlinear absorption on open aperture Z-scan measurements is discussed.

2. Coordinate transformation of beam propagation

The particular coordinate transformation is based on the vacuum behaviour of a focused Gaussian beam.[12] Some other authors used similar systems[13,14] to enhance the computational speed and accuracy in analyses of laser resonator cavity and beam propagating in atmosphere.

The basic concept of the coordinate transformation is to calculate the amplitude changes induced in the vacuum rather than to calculate the complete amplitude. Therefore, this procedure removes the necessity for sampling the high frequency oscillation in the phase induced by focusing. The coordinate transformation alters the independent variables so that the dependent variables take a different functional form, but are numerically identical to the untransformed amplitude at equivalent points in space in the two coordinate systems.

The paraxial wave equation including linear and nonlinear absorption in the Fresnel approximation can

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†Corresponding author. E-mail: jjtian@nankai.edu.cn

be written as

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial E}{\partial r}\right) - 2\mathrm{i}k\frac{\partial E}{\partial z} - \mathrm{i}k\alpha_1 E - \mathrm{i}k\alpha_m |E|^{2(m-1)} E + \frac{2n_2k^2}{n_0}|E|^2 E = 0, \quad (1)$$

where k is the wave vector, n_0 is the linear refractive-index, α_m is the m-photon absorption coefficient and n_2 is the nonlinear refractive-index coefficient.

Assuming a TEM_{00} Gaussian beam travelling in the $+z$ direction, we can write the electric field E as follows:

$$E(r,z,t) = E_0(t)\frac{w_0}{w(z)}\exp\left(-\frac{r^2}{w^2(z)} - \frac{\mathrm{i}kr^2}{2R(z)} + \mathrm{i}\tan^{-1}(z/z_0)\right), \quad (2)$$

where $w(z) = w_0\left(1 + z^2/z_0^2\right)^{1/2}$ is the beam radius, $R(z) = z\left(1 + z_0^2/z^2\right)$ is the radius of curvature of the wave-front at z, w_0 is the radius of beam in focus and $z_0 = kw_0^2/2$ is the Rayleigh length of the beam. $E_0(t)$ denotes the radiation electric field at the focus and contains the temporal envelope of laser pulse.

As explained in Ref.[15], the existence of the quadratic term in the imaginary part of the exponential will cause a high-frequency oscillation in the amplitude for $r > w(z)$. This can exceed the sampling frequency of the computational mesh. Therefore, a phase change shall be made to remove this term. Additionally, the amplitude will increase as $z \to 0$ due to the existence of the term $w_0/w(z)$. Identically, it is desirable to remove this factor as well. As the beam focuses and becomes smaller, the mathematical mesh should shrink at the same rate to model the transverse distribution accurately. These considerations lead to the following sequences of coordinate transformations:

$$E(\bar{r},\bar{z}) \equiv E_0(t)C(\bar{r},\bar{z})A(\tilde{r},\tilde{z}), \quad (3a)$$

$$C(\bar{r},\bar{z}) = \frac{w_0}{w(\bar{z})}\exp\left[-\frac{\mathrm{i}k\bar{r}^2}{2R(\bar{z})} + \mathrm{i}\tan^{-1}(\bar{z})\right], \quad (3b)$$

$$\frac{\mathrm{d}\tilde{z}}{\mathrm{d}\bar{z}} = \frac{1}{1+\bar{z}^2} = \frac{1}{D}, \quad (3c)$$

where $\bar{z} = z/z_0$, $\bar{r} = r/w_0$, and $\tilde{r} = \bar{r}/(1+\bar{z}^2)^{1/2}$. The phase of function $A(\tilde{r},\tilde{z})$ changes very slowly with $\tilde{r}$. The quadratic term in the imaginary part of the exponential in Eq.(3b), which will cause a high-frequency oscillation in the amplitude for $r > w(z)$, and the term $w_0/w(z)$ have been included in the function $C(\bar{r},\bar{z})$. Function $C(\bar{r},\bar{z})$ can be solved analytically. If the function $A(\tilde{r},\tilde{z})$ is obtained, we can obtain the electric field through Eq.(3a). Rewriting Eq.(1) by use of the function $A(\tilde{r},\tilde{z})$ in these new coordinates, the following equations can be obtained:

$$\frac{1}{\tilde{r}}\frac{\partial}{\partial\tilde{r}}\left(\tilde{r}\frac{\partial A}{\partial\tilde{r}}\right) + 4\left(1-\tilde{r}^2\right)A - 4\mathrm{i}\frac{\partial A}{\partial\tilde{z}} - p(A)A = 0, \quad (4a)$$

$$p(A) = 2\mathrm{i}\sum_{m=1}^{n}\bar{\alpha}_m |A|^{2(m-1)}\frac{A}{D^{m-2}}, \quad (4b)$$

where $\bar{\alpha}_m = \alpha_m I_0^{(m-1)} z_0$ is the nonlinear absorption at focus for the nonlinear medium with one Rayleigh length.

For simplicity, we ignore the coupling between nonlinear absorption and nonlinear refraction, and set $n_2 = 0$. For the case with $p(A) = 0$, which corresponds to the beam propagation in vacuum, the transformed field has the following form:

$$A(\tilde{r},\tilde{z}) = \exp(-\tilde{r}^2). \quad (5)$$

This means that the form of the amplitude in vacuum is unchanged as the beam propagates in this coordinate system. Therefore, the use of the transformations in Eqs.(4) can reduce significantly the size of the data arrays and the computational time, since a fewer points can express the radial field distribution and the propagation distance is shrunken greatly. Numerical computing has shown that the numerical computing efficiency for Eqs.(4) has been enhanced two order of magnitude as against that for Eq.(1).[15]

For the case of a thick medium, the numerical computing shows that the change of the shape of electric field $A(\tilde{r},\tilde{z})$, which is induced by nonlinear absorption, can be ignored. We can define the normalized intensity as follows:

$$I_N(\tilde{r},\tilde{z}) = \left|\frac{A(\tilde{r},\tilde{z})}{A(0,\tilde{z})}\right|^2. \quad (6)$$

The change induced by nonlinear absorption is only the amplitude of normalized intensity when nonlinear refraction is absent or the coupling between refraction and absorption can be ignored. The normalized intensities for several positions inside the medium are given in Fig.1 by solving Eq.(4a) numerically.[15] We can see from Fig.1 that the change of shape of normalized intensity is very small. This implies that the beam shape is almost unchanged and there exists effectively uniform attenuation across the radial dimension of the beam. The beam is most strongly absorbed along the

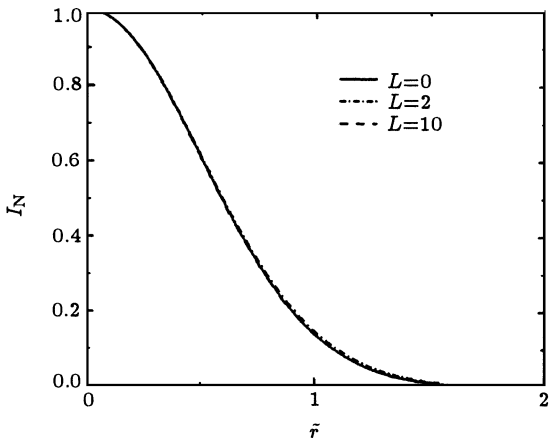


Fig.1. The normalized intensities for some positions in the medium. The unit of medium length is Rayleigh length z_0. The other parameters are $\bar{\alpha}_2 = 1$, and linear absorption coefficient $\bar{\alpha}_1 = 1$.

axis where the irradiance is highest across radial dimension, presumably because diffraction redistributes off-axis light towards the axis for compensating the central loss. We define the scaled peak intensity (SPI), $I_S(0,\bar{z}) = |A(0,\bar{z})|^2$. $I_S(0,\bar{z}=x) = 1$ is the SPI in the front surface of medium, x is the position of the front surface of medium. So the scaled intensity in medium can be expressed as

$$I_{\mathrm{SI}}(\tilde{r},\bar{z}) = I_S(0,\bar{z})\exp\left(-2\tilde{r}^2\right). \tag{7}$$

Because the shape of electric field $A(\tilde{r},\bar{z})$ keeps unchanged, the first and second terms on the left-hand side of Eq.(4a) counteract each other. Inserting Eq.(7) into Eq.(4a) and completing the integration about cross section of beam, we find that the SPI attenuates according to

$$\frac{\mathrm{d}I_{\mathrm{S}}(0,\bar{z})}{\mathrm{d}\bar{z}} = -\bar{\alpha}_1 I_{\mathrm{S}}(0,\bar{z}) - \bar{\alpha}_m \frac{I_{\mathrm{S}}^m(0,\bar{z})}{mD^{(m-1)}}. \tag{8}$$

Equation (8) can be solved exactly and the solution can be written as

$$I_{\mathrm{S}}(0,\bar{z}) = \frac{\exp[-\bar{\alpha}_1(\bar{z}-x)}{\left(1+\bar{\alpha}_m \times \dfrac{(m-1)}{m} \times L^{\mathrm{equ}}(\alpha_1,\bar{z},x)\right)^{\frac{1}{m-1}}}, \tag{9a}$$

$$L^{\mathrm{equ}}(\bar{\alpha}_1,\bar{z},x) = \int_x^{\bar{z}} \left[\frac{\exp[-\bar{\alpha}_1(\bar{z}'-x)]}{(1+\bar{z}'^2)}\right]^{(m-1)} \mathrm{d}\bar{z}', \tag{9b}$$

where $m \geq 2$.

3. Z-scan analysis and results

Using Eq.(7) and Eqs.(9), we can obtain the normalized transmittance of open-aperture Z-scan by the ratio of the beam intensity in the front surface to that in the exit interface

$$\begin{aligned} T(x,l) =& \frac{I_{\mathrm{S}}(\bar{\alpha}_1,\bar{\alpha}_m,l)}{I_{\mathrm{S}}(\bar{\alpha}_1,\bar{\alpha}_m=0,l)} \\ =& \frac{1}{\left(1+\bar{\alpha}_m \times \dfrac{(m-1)}{m} \times L^{\mathrm{equ}}(\alpha_1,l,x)\right)^{\frac{1}{m-1}}}. \end{aligned} \tag{10}$$

The formula is in a general form for multiphoton absorption case.

For the two-photon absorption case, Eq.(10) can be reduced to

$$T(x,l) = \frac{1}{\left(1+\bar{\alpha}_2 \times \dfrac{1}{2} \times L^{\mathrm{equ}}(\alpha_1,l,x)\right)}, \tag{11a}$$

where

$$L^{\mathrm{equ}}(\bar{\alpha}_1,l,x) = \int_x^{x+l} \frac{\exp[-\bar{\alpha}_1(\bar{z}'-x)]}{(1+\bar{z}'^2)} \mathrm{d}\bar{z}'. \tag{11b}$$

If the linear absorption is absent, $\bar{\alpha}_1 = 0$, we can complete the integration of Eq.(11b) and obtain the following expression:

$$L^{\mathrm{equ}}(\bar{\alpha}_1 = 0,l,x) = \tan^{-1}(x+l) - \tan^{-1}(x), \tag{12}$$

which is the distance between the front surface and exit surface of medium in this new coordinate system and we can call it equivalent medium length. The relation between equivalent medium lengths, medium length and the position of medium is shown in Fig.2. If the linear absorption coefficient exists, the equivalent medium length can be expressed as Eq.(11b). The relation between the equivalent medium length and the linear absorption coefficient is shown in Fig.3 and the real medium length is $4z_0$. We can see that the linear absorption coefficient affects the equivalent medium length and the position of equivalent medium length. Therefore, the linear absorption coefficient must also affect the magnitude of absorption dip and the position of absorption dip. The relation between the absorption dip of normalized transmittance and the linear absorption coefficient is shown in Fig.4, and the real medium length is $4z_0$. We can see that the absorption dip decreases as linear absorption coefficient increases. This is because that the equivalent medium

length decreases as the linear absorption coefficient increases. The relation between the position of absorption dip of normalized transmittance and the linear absorption coefficient is shown in Fig.5 and the real medium length is the same as that in Fig.4. We can see that the position of absorption dip moves towards the front surface of medium as the linear absorption coefficient increases. This is because the equivalent medium length decreases as linear absorption coefficient increases.

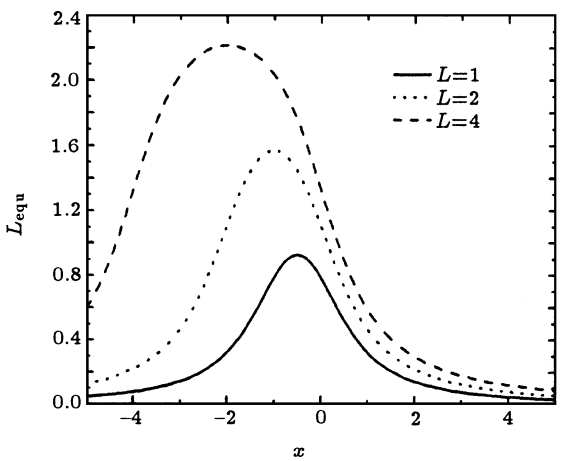


Fig.2. Equivalent medium length L_{equ} versus the position of medium x. The unit of medium length L is Rayleigh length z_0.

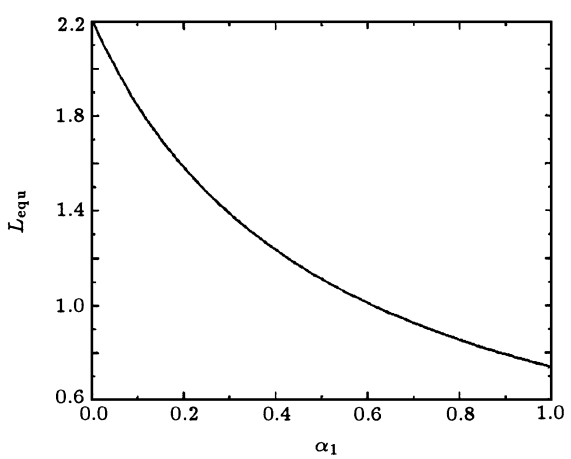


Fig.3. The equivalent medium length versus the linear absorption coefficient.

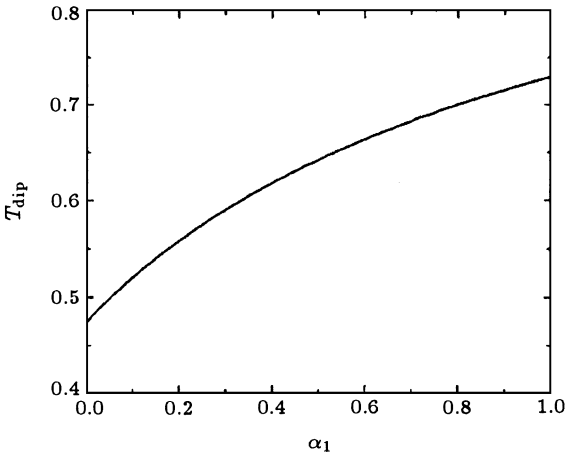


Fig.4. The absorption dip of normalized transmittance versus the linear absorption coefficient. The real medium length is $4z_0$, and $\bar{\alpha}_2 = 1$.

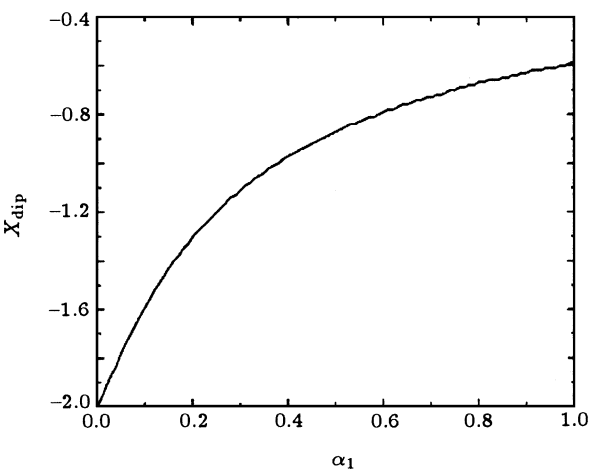


Fig.5. The position of the absorption dip versus the linear absorption coefficient. The real medium length is $4z_0$.

4. Conclusion

In this paper, we have analysed the open-aperture Z-scan with multiphoton absorption by coordinate transformation. The approximate closed-form solution is obtained. The coupling between linear absorption and nonlinear absorption is also analysed in detail. Results show that not only the magnitude of the absorption dip, but also the position of absorption dip of the normalized transmittance of open-aperture Z-scan is affected seriously by linear absorption coefficient.

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