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THEORETICAL ANALYSIS OF OPEN APERTURE REFLECTION Z-SCAN ON MATERIALS WITH HIGH-ORDER OPTICAL NONLINEARITIES

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Abstract

We present a theoretical analysis of open aperture reflection Z-scan in nonlinear media with third-, fifth-, and higher-order nonlinearities. A general analytical expression for the normalized reflectance when third-, fifth- and higher-order optical nonlinearities are excited is derived and its consequences on RZ-scan in media with high-order nonlinearities are discussed. We show that by performing RZ-scan experiments at different incident intensities it is possible to put in evidence the excitation of different order nonlinearities in the medium. Their contributions to the overall nonlinear response can be discriminated by using formulas derived by us. A RZ-scan numerical simulation using these formulas and data taken from literature, measured by another method for the third-, fifth-, and seventh-order nonlinear refractive indices of As_2S_3 chalcogenide glass, is performed.

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1. Introduction

The Z-scan method allows the measurement of the magnitude and the sign of the optical nonlinearities [1]. It is based on the dependence of the nonlinear refractive index / nonlinear absorption coefficient on the local beam intensity in the sample. In the Z-scan method, the investigated sample is moving along the direction of a focused incident beam, passing through the focal plane of the focusing lens and the phase and / or amplitude distortions of the transmitted / reflected beams are monitored in the far field.

The optical nonlinearities are quantitatively measured analyzing the beam transmitted by the sample, in the transmission Z-scan (TZ-scan) method [1, 2] and the beam reflected by the sample, in the reflection Z-scan (RZ-scan) method [3, 4].

TZ-scan is used for the measurement of bulk nonlinear optical properties of transparent media. The nonlinear refraction is measured in closed-aperture TZ-scan configuration while the nonlinear absorption in open-aperture TZ-scan.

RZ-scan is used for the investigation of highly absorbing nonlinear media, when TZ-scan cannot be used, or for the study of surface nonlinear optical properties of transparent media. In open aperture RZ-scan configuration, the refractive nonlinearities are measured by monitoring the total change of the reflection coefficient. These nonlinearities are responsible for the amplitude changes of the reflected radiation [5-9]. In closed aperture RZ-scan configuration, the absorptive nonlinearities, responsible for the phase changes in the reflected beam, are measured [4, 8].

In this paper, we will refer to the open aperture RZ-scan, only. A schematic of the open aperture RZ-scan experimental configuration and the typical Z-scan traces for nonlinear media with positive / negative refractive third-order nonlinearities are shown in the Fig. 1.

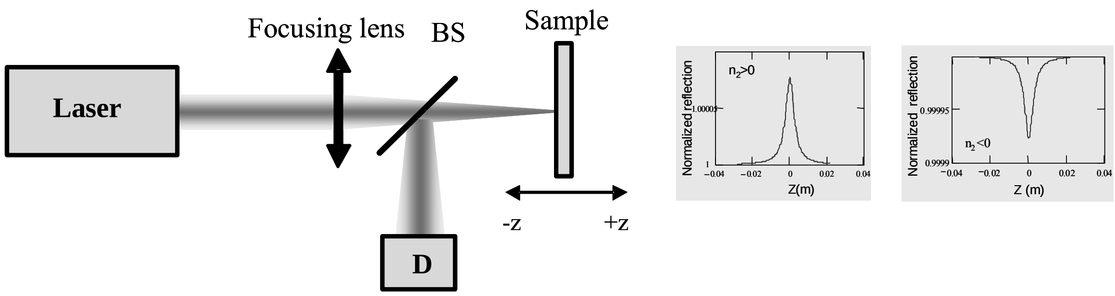


Fig. 1. Open aperture RZ-scan configuration and typical RZ-scan traces for nonlinear media with positive / negative refractive third-order nonlinearities

When the sample is far from the focal plane of the focusing lens, the intensity of the incident light is low and the reflection coefficient R_0 of the sample is not changing (nonlinear effects are negligible). Around the focal plane, the incident intensities are much higher and the nonlinear refraction effects occur, changing the magnitude of the reflection coefficient. For a positive nonlinearity (nonlinear refractive index $n_2 > 0$), the reflection coefficient $R(z)$ increases when the sample is approaching the focal plane, due to the increasing intensity, and decreases after passing through the focal plane, going down to the linear value, R_0. For a sample with a negative nonlinearity ($n_2 < 0$), the $R(z)$ dependence has a valley around the focal plane.

The fifth-, seventh- and higher-order optical nonlinearities are usually much weaker than the third-order one. At low laser intensities their contribution to the nonlinear response of the material is negligible and it is not considered in Z-scan experiments. But in Z-scan experiments performed with ultrashort laser pulses, at high intensities, the contribution of the fifth-, seventh-, and even higher-order nonlinearities to the optical response of the excited medium can be significant [10-17].

The nonlinear optical response of materials in which fifth-, and higher-order nonlinearities are excited was theoretically analyzed and modeled for TZ-scan [2, 15, 18-20].

For RZ-scan, to the best of our knowledge, a theoretical modeling of the effect of fifth- and higher-order nonlinearities on the nonlinear reflectance does not yet exist. The normalized reflectance curve obtained in a RZ-scan experiment contains the information about all involved nonlinearities. The discrimination between the contributions of third-, fifth- and higher-order nonlinearities to the overall nonlinear response is difficult because of the lack of related theories [21]. A specific expression for each order of the nonlinearities excited in the medium must be used to correctly fit the experimental RZ-scan curves.

In this work we analyze the contribution of high-order nonlinearities on open-aperture RZ-scan signal and we derive a general analytical expression for the normalized reflectance when third-, fifth- and higher-order optical nonlinearities are excited. The consequences of this expression for RZ-scan when high-order nonlinearities are excited, are discussed. We show that by performing RZ-scan experiments at different incident intensities it is possible to put in evidence the excitation of different order nonlinearities in the medium. Their contribution to the overall nonlinear response can be discriminated by using formulas derived by us. The results of our theoretical model for RZ-scan are illustrated by a numerical simulation of the normalized reflectance on As_2S_3 chalcogenide glass, in which the magnitude of the third-, fifth-, and seventh-order nonlinearities have been estimated by spectrally resolved two-beam coupling.

2. Theoretical analysis of open aperture RZ-scan on materials with high-order optical nonlinearities

For quantitative measurement of the refractive nonlinearities in open aperture RZ-scan normalized reflectance, defined as the ratio between the reflectance with and without the nonlinear effect, is used [5]:

$$R(z,\theta)=\frac{\int_0^\infty |r(\theta)|^2 I(\rho,z)\rho d\rho}{\int_0^\infty |r_0(\theta)|^2 I(\rho,z)\rho d\rho} \quad , \tag{1}$$

where ρ and z are the radial and axial coordinates, $r(\theta)$ is the amplitude reflection coefficient, θ is the angle of incidence of the focused laser beam on the sample surface, $I(\rho, z)$ is the incident beam intensity.

At normal incidence, the amplitude reflection coefficient $r(0)$ of a medium with complex refractive index $\tilde{n}$, is given by the Fresnel expression:

$$r(0)=\frac{\tilde{n}-1}{\tilde{n}+1} \quad , \tag{2}$$

where $\tilde{n}=n+ik$ is the complex refractive index, in which n is the real refractive index and k is the extinction coefficient.

We consider nonlinear materials with $n >> k$. In this case $\tilde{n} \approx n$ and Eq. (2) becomes:

$$r(0)\equiv r=\frac{n-1}{n+1} \quad . \tag{3}$$

In the linear regime, the linear amplitude reflection coefficient is:

$$r_0(0)\equiv r_0=\frac{n_0-1}{n_0+1} \quad , \tag{4}$$

with n_0 the linear refractive index.

The total refractive index n of an illuminated nonlinear material can be written as:

$$n = n_0 + \Delta n(I) \ , \tag{5}$$

in which $\Delta n(I)$ is the light induced change of the refractive index that can be written as $\Delta n(I) = n_2 I$, for a medium with third-order optical nonlinearity only, and as:

$$\Delta n(I) = n_2 I + n_4 I^2 + n_6 I^3 \ , \tag{6}$$

for a medium in which third-, fifth- and seventh-order nonlinearities are excited. The coefficients n_2, n_4 and n_6 are the nonlinear refractive indices corresponding to the third-, fifth-, and seventh-order optical nonlinearity, respectively.

Considering the amplitude reflection coefficients, in the nonlinear reflection regime, r, and in the linear one, r_0, from Eq. (3) and Eq. (4), respectively, the RZ-scan normalized reflectance when the light beam is incident at normal incidence ($\theta = 0$) on the sample surface becomes:

$$R(z) = \frac{\int_0^\infty |r|^2 I(\rho, z)\rho d\rho}{\int_0^\infty |r_0|^2 I(\rho, z)\rho d\rho} \ , \tag{7}$$

We consider the case of a nonlinear medium in which third-, fifth- and seventh-order nonlinearities are excited. In this case, the refractive index can be written as:

$$n = n_0 + n_2 I + n_4 I^2 + n_6 I^3 \ . \tag{8}$$

For a small change of the refractive index, $\Delta n << n_0$, the reflection coefficient r can be expanded in first order and written as [5]:

$$r = r_0 + \frac{\partial r}{\partial n}\Delta n \ . \tag{9}$$

The derivative $\frac{\partial r}{\partial n}$ is:

$$\frac{\partial r}{\partial n} = \frac{2}{(n+1)^2} \cong \frac{2}{(n_0+1)^2} \ . \tag{10}$$

With Eqs. (9), (10) and (4), the normalized reflectance, $R(z)$ from Eq. (7), can be written as:

$$R(z) = \frac{\int_0^\infty \left(r_0 + \frac{\partial r}{\partial n}\Delta n \right)^2 I(\rho, z)\rho d\rho}{\int_0^\infty r_0^2 I(\rho, z)\rho d\rho} \approx \frac{\int_0^\infty \left(r_0^2 + 2r_0 \frac{\partial r}{\partial n}\Delta n \right) I(\rho, z)\rho d\rho}{\int_0^\infty r_0^2 I(\rho, z)\rho d\rho} = 1 + \frac{4}{n_0^2 - 1} \cdot \frac{\int_0^\infty \Delta n \cdot I(\rho, z)\rho d\rho}{\int_0^\infty I(\rho, z)\rho d\rho} \tag{11}$$

In Eq. (11), the term $\left(\frac{\partial r}{\partial n}\right)^2 (\Delta n)^2$ was neglected due to the small light induced change of the refractive index, $\left(\frac{\partial r}{\partial n}\right)^2 (\Delta n)^2 << 2r_0 \frac{\partial r}{\partial n}\Delta n$.

Taking into consideration Eq. (6) for the light induced change of the refractive index, Eq. (11) becomes:

$$R(z) = 1 + \frac{4n_2}{n_0^2 - 1} \cdot \frac{\Im_2}{\Im_1} + \frac{4n_4}{n_0^2 - 1} \cdot \frac{\Im_3}{\Im_1} + \frac{4n_6}{n_0^2 - 1} \cdot \frac{\Im_4}{\Im_1} \ , \tag{12}$$

where:

$$\Im_1 = \int_0^\infty I(\rho, z)\rho d\rho \ , \ \ \Im_2 = \int_0^\infty I^2(\rho, z)\rho d\rho \ , \ \ \Im_3 = \int_0^\infty I^3(\rho, z)\rho d\rho \ , \ \ \Im_4 = \int_0^\infty I^4(\rho, z)\rho d\rho \ . \tag{13}$$

The intensity $I(\rho, z)$ of a Gaussian beam is given by:

$$I(\rho, z) = I_0 \left(\frac{w_0}{w(z)} \right)^2 \exp\left[\frac{-2\rho^2}{w(z)^2} \right] \ , \tag{14}$$

where w_0 is the beam waste,

$$w(z) = w_0 \left[1 + (z/z_0)^2 \right]^{\frac{1}{2}} \ , \tag{15}$$

$I_0 = I(0,0)$ is the peak intensity and z_0 is the Rayleigh length, $z_0 = \pi w_0^2 / \lambda$. Introducing Eqs. (14)-(15) in the integrals expressed in Eq. (13), these integrals can be written as:

$$\Im_1 = \frac{I_0}{1 + (z/z_0)^2} \cdot \Im_1^{'} \ , \ \Im_2 = \frac{I_0^2}{\left[1 + (z/z_0)^2 \right]^2} \cdot \Im_2^{'} \ , \ \Im_3 = \frac{I_0^3}{\left[1 + (z/z_0)^2 \right]^3} \cdot \Im_3^{'} \ , \ \Im_4 = \frac{I_0^4}{\left[1 + (z/z_0)^2 \right]^4} \cdot \Im_4^{'} \ , \tag{16}$$

where $\Im_1^{'}$, $\Im_2^{'}$, $\Im_3^{'}$, $\Im_4^{'}$ are:

$$\Im_1^{'} = \int_0^\infty \rho \cdot \exp\left(-\frac{2\rho^2}{w^2(z)} \right) \cdot d\rho \ , \quad \Im_2^{'} = \int_0^\infty \rho \cdot \exp\left(-\frac{4\rho^2}{w^2(z)} \right) \cdot d\rho \ ,$$

$$\Im_3^{'} = \int_0^\infty \rho \cdot \exp\left(-\frac{6\rho^2}{w^2(z)} \right) \cdot d\rho \ , \quad \Im_4^{'} = \int_0^\infty \rho \cdot \exp\left(-\frac{8\rho^2}{w^2(z)} \right) \cdot d\rho \ , \tag{17}$$

respectively. After integration, we obtain for $\Im_1^{'}$, $\Im_2^{'}$, $\Im_3^{'}$, $\Im_4^{'}$:

$$\Im_1^{'} = \frac{w^2(z)}{4} \ , \quad \Im_2^{'} = \frac{w^2(z)}{8} \ , \quad \Im_3^{'} = \frac{w^2(z)}{12} \ , \quad \Im_4^{'} = \frac{w^2(z)}{16} \ . \tag{18}$$

The normalized reflectance, $R(z)$, from Eq. (12) can now be written as:

$$R(z) = 1 + \frac{2n_2 I_0}{n_0^2 - 1} \cdot \frac{1}{1 + (z/z_0)^2} + \frac{\frac{4}{3} n_4 I_0^2}{n_0^2 - 1} \cdot \frac{1}{\left[1 + (z/z_0)^2 \right]^2} + \frac{n_6 I_0^3}{n_0^2 - 1} \cdot \frac{1}{\left[1 + (z/z_0)^2 \right]^3} \ . \tag{19}$$

We can write this formula as:

$$\begin{aligned} R(z) &= 1 + \frac{4n_2}{n_0^2 - 1} \cdot \frac{1}{2} \cdot \frac{I_0}{1 + (z/z_0)^2} + \frac{4n_4}{n_0^2 - 1} \cdot \frac{1}{3} \cdot \frac{I_0^2}{\left[1 + (z/z_0)^2 \right]^2} + \frac{4n_6}{n_0^2 - 1} \cdot \frac{1}{4} \cdot \frac{I_0^3}{\left[1 + (z/z_0)^2 \right]^3} = \\ &= 1 + \frac{4}{n_0^2 - 1} \left\{ \frac{1}{2} \cdot n_2 \cdot \left[\frac{I_0}{1 + (z/z_0)^2} \right]^1 + \frac{1}{3} \cdot n_4 \cdot \left[\frac{I_0}{1 + (z/z_0)^2} \right]^2 + \frac{1}{4} \cdot n_6 \cdot \left[\frac{I_0}{1 + (z/z_0)^2} \right]^3 \right\} \end{aligned} \tag{20}$$

This is the normalized reflectance in open-aperture RZ-scan when the contributions of the third-, fifth- and seventh-order nonlinearities to the overall nonlinear response are considered.

We will denote by $R_3(z)$ the normalized reflectance when we take into consideration the contribution of third-order nonlinearity only, by $R_{3\text{-}5}(z)$ the normalized reflectance when we take into consideration the contribution of third- and fifth-order nonlinearities, and by $R_{3\text{-}5\text{-}7}(z)$ the normalized reflectance when we take into consideration the contribution of third-, fifth- and seventh-order nonlinearities. With these notations we have:

$$R_3(z) = 1 + \Delta R_3(z), \quad R_{3-5}(z) = 1 + \Delta R_3(z) + \Delta R_5(z), \quad R_{3-5-7}(z) = 1 + \Delta R_3(z) + \Delta R_5(z) + \Delta R_7(z).$$

$\Delta R_3(z)$, $\Delta R_5(z)$, $\Delta R_7(z)$ are the changes of the normalized reflectance due to the contributions of the third-, fifth- and seventh-order nonlinearities, respectively:

$$\Delta R_3(z) = \frac{4n_2}{n_0^2-1}\cdot\frac{1}{2}\cdot\frac{I_0}{1+(z/z_0)^2} = \frac{4}{n_0^2-1}\cdot\frac{1}{2}\cdot\frac{\Delta n_3}{1+(z/z_0)^2}\ , \tag{21}$$

$$\Delta R_5(z) = \frac{4n_4}{n_0^2-1}\cdot\frac{1}{3}\cdot\frac{I_0^2}{\left[1+(z/z_0)^2\right]^2} = \frac{4}{n_0^2-1}\cdot\frac{1}{3}\cdot\frac{\Delta n_5}{\left[1+(z/z_0)^2\right]^2}\ , \tag{22}$$

$$\Delta R_7(z) = \frac{4n_6}{n_0^2-1}\cdot\frac{1}{4}\cdot\frac{I_0^3}{\left[1+(z/z_0)^2\right]^3} = \frac{4}{n_0^2-1}\cdot\frac{1}{4}\cdot\frac{\Delta n_7}{\left[1+(z/z_0)^2\right]^3}\ , \tag{23}$$

where Δn_3, Δn_5, Δn_7 are the changes of the refractive index produced by the third-, fifth-, and seventh-order nonlinearity, respectively.

The formula for the normalized reflectance $R_3(z)$, when it is considered the third-order nonlinearity only, is the same as those previously obtained for open-aperture RZ-scan [4, 5, 22] which show that the normalized reflectance $R(z)$ is proportional to the (small) change of the real part of refractive index, Δn.

We can write Eq. (20) in a more compact and general form taking into account the contribution of higher than seventh-order nonlinearities, as well. We shall assign the order of the nonlinearity with the index l = 3, 5, 7,…, for the nonlinearities of the 3rd, 5th, 7th, …, order respectively, or with another index, p, related to l by the relation $l=2p+1$. The p index has values p = 1, 2, 3,…, for the 3rd, 5th, 7th, …, order, respectively.

By using the index l, the normalized reflectance can be written as:

$$R(z) = 1 + \frac{4}{n_0^2-1}\sum_{l=3,5,7,\ldots}\left\{\frac{2}{l+1}\cdot n_{l-1}\cdot\left[\frac{I_0}{1+(z/z_0)^2}\right]^{\frac{l-1}{2}}\right\}\ , \tag{24}$$

whereas using the index p, we can write the normalized reflectance as:

$$R(z) = 1 + \frac{4}{n_0^2-1}\sum_{p=1,2,3,\ldots}\left\{\frac{1}{p+1}\cdot n_{2p}\cdot\left[\frac{I_0}{1+(z/z_0)^2}\right]^{p}\right\}\ . \tag{25}$$

Equations (24), (25) are general formulas for the normalized reflectance in open-aperture RZ-scan when it is considered not only the third-order nonlinearity, but also the contribution of higher-order nonlinearities.

From Eqs. (24), (25) it is possible to see that the excitation of different order nonlinearities does not change only the magnitude of the normalized reflectance, but its dependence on z, too. The contribution of higher-order nonlinearity to the magnitude of the overall RZ-scan signal becomes larger when the sample is approaching the focal plane, due to the dependence of normalized reflectance on intensity I_0 at a higher power. On the other hand, the RZ-scan curve produced when considering the effect of a single nonlinearity only becomes sharper when the order of the nonlinearity increases, due to the factor $1/(1+z/z_0)^2$ at higher powers. These are consequences of the increased spatial localization, in regions with higher intensities, of higher-order nonlinearities excitation. We have to mention also the possible appearance in the RZ-scan curve of a sharp local peak (valley) on a larger peak (valley) as a result of nonlinear refractive indices with the same or with opposite signs corresponding to excited nonlinearities of different orders that contribute to the overall nonlinear response of the material.

The derived formulas for the normalized reflectance can be used for characterization of nonlinear optical processes when a single nonlinearity of third-, fifth-, or higher-order is excited separately, or when several nonlinearities of different orders are excited simultaneously. They are suitable for the analysis of saturating nonlinearities as well, when the change of the refractive index, $\Delta n(I)$ can be expressed as [10, 23]:

$$\Delta n(I) = \frac{n_2 I}{1 + I/I_{sat}} \quad , \tag{26}$$

where I_{sat} is the saturation intensity. The change of the refractive index can be expanded in the perturbative form as [10]:

$$\Delta n(I) = n_2 I - \frac{n_2}{I_{sat}} I^2 + \frac{n_2}{(I_{sat})^2} I^3 - \ldots \equiv n_2 I + n_4 I^2 + n_6 I^3 + \ldots \ . \tag{27}$$

From Eq. (27) it is easy to see that the overall change of the refractive index can be expressed as a summation of the contributions due to the successive orders nonlinearities. The contribution of higher-order terms increases when I increases.

Now we will analyze the dependence of the RZ-scan peak or valley amplitude (corresponding to z=0) on the incident laser intensity, I_0. Considering the expression for the normalized reflectance, $R_3(z)$, when only the third-order nonlinearity is excited, the amplitude of the peak (or valley) in RZ-scan dependence is given by:

$$\Delta R_3 \equiv \Delta R_3(0) = R_3(0) - R_3(\mp\infty) = \frac{2n_2}{n_0^2 - 1} \cdot I_0 = A_3 \cdot I_0 \ , \tag{28}$$

where $A_3 = 2n_2/(n_0^2 - 1)$. Thus, the dependence on the laser intensity, I_0, of the peak (valley) amplitude $\Delta R_3(I_0) = A_3 \cdot I_0$ is a linear function of I_0. Alternatively, $\Delta R_3(I_0)/I_0 = A_3$ is a constant function of I_0.

When both third- and fifth-order nonlinearities are excited, the amplitude of the peak (or valley) in the RZ-scan dependence, $R_{3\text{-}5}(z)$ is given by:

$$\Delta R_{3-5} = \frac{2n_2}{n_0^2 - 1} \cdot I_0 + \frac{(4/3)n_4}{n_0^2 - 1} \cdot I_0^2 = A_3 \cdot I_0 + B_{3-5} I_0^2 \ , \tag{29}$$

where $B_{3-5} = (4/3)n_4/(n_0^2 - 1)$. Thus, in this case, $\Delta R_{3-5}(I_0) = A_3 \cdot I_0 + B_{3-5} \cdot I_0^2$ is a parabolic function of I_0. Alternatively, $\Delta R_{3-5}(I_0)/I_0 = A_3 + B_{3-5} I_0$, is a linear function of I_0.

When third-, fifth- and seventh-order nonlinearities are excited, the amplitude of the peak (or valley) in the RZ-scan dependence, $R_{3\text{-}5\text{-}7}(z)$, is given by:

$$\Delta R_{3-5-7} = \frac{2n_2}{n_0^2 - 1} \cdot I_0 + \frac{(4/3)n_4}{n_0^2 - 1} \cdot I_0^2 + \frac{n_6}{n_0^2 - 1} \cdot I_0^3 = A_3 \cdot I_0 + B_{3-5} I_0^2 + C_{3-5-7} I_0^3 \tag{30}$$

where $C_{3-5-7} = n_6 / (n_0^2 - 1)$. Thus, in this case, $\Delta R_{3-5-7}(I_0) = A_3 \cdot I_0 + B_{3-5} I_0^2 + C_{3-5-7} I_0^3$ is a third-order polynomial function of I_0, and $\Delta R_{3-5-7} / I_0 = A_3 + B_{3-5} I_0 + C_{3-5-7} I_0^2$, is a parabolic function of I_0.

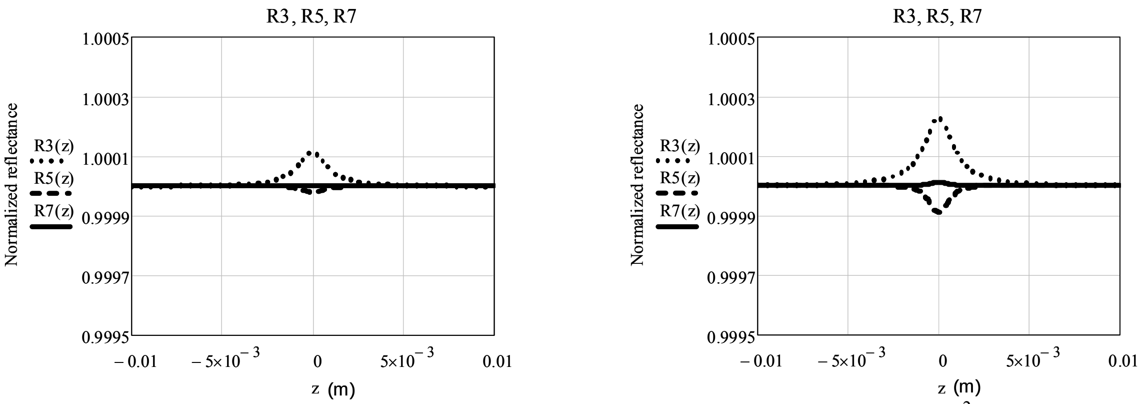
Thus, RZ-scan experiments carried out at different incident intensities allow us to put in evidence the presence, or absence, of different order nonlinearities in the investigated material, by analyzing the I_0 dependence of the normalized reflectance peak (valley) amplitude. Eqs. (28) – (30), or similar equations, obtained for other values of fixed z, $z\neq 0$, can be used to fit the normalized reflectance in intensity scan (I-scan) experiments, performed in the focal plane, z=0, or in other planes, $z\neq 0$.

3. Numerical simulation of open aperture RZ-scan

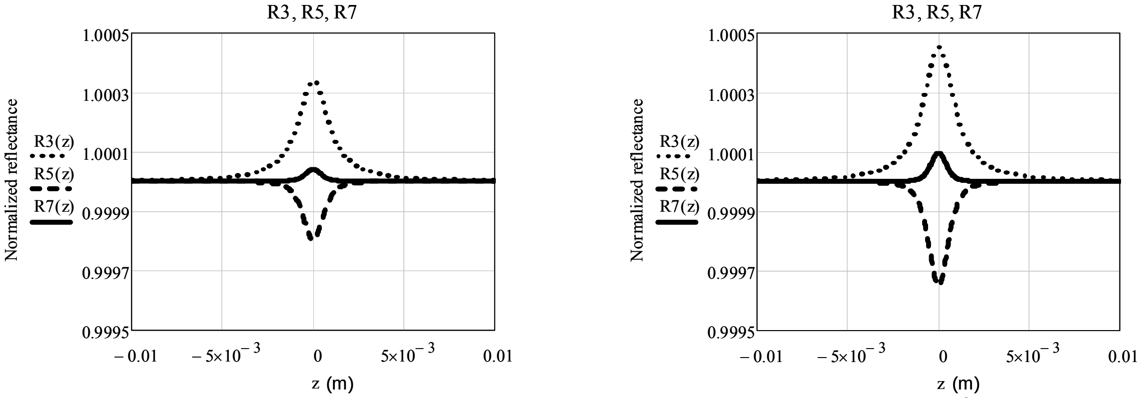
In order to illustrate our RZ-scan analytical results, we performed a numerical simulation of the normalized reflectance on As_2S_3 chalcogenide glass. In this material third-, fifth- and seventh-order nonlinearities have been estimated by spectrally resolved two-beam coupling performed at the pump wavelength of 790 nm with 100 fs pulses [10]. The values of n_2, n_4 and n_6 nonlinear refractive indices are as follows: n_2 = 2.7x10^{-13} cm^2/W, n_4 = -7.8x10^{-23} cm^4/W^2, n_6 = 7.2x10^{-33} cm^6/W^3. The measurements performed at different intensities of the pump beam revealed a saturation trend of the nonlinear response with the saturation intensity I_{sat} ~ 3GW/cm^2.

In our simulation, we considered the refractive index of As_2S_3 as n_0=2.4 and the Rayleigh length in the RZ-scan configuration as z_0 = 1 mm. The incident intensity, I_0, was increased up to the value of 4GW/cm^2, larger than the saturation intensity I_{sat} in As_2S_3.

Fig. 2 shows the normalized reflectances R_3, R_5, R_7 if the corresponding nonlinearities of the third-, fifth-, and seventh-order, respectively, would be excited separately. The RZ-curve becomes sharper as the order of the excited nonlinearity increases.



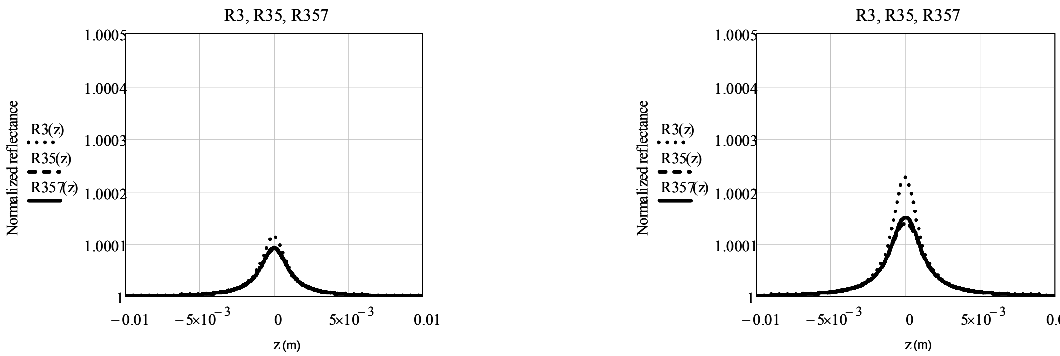
I_0=1 GW/cm^2 I_0=2 GW/cm^2



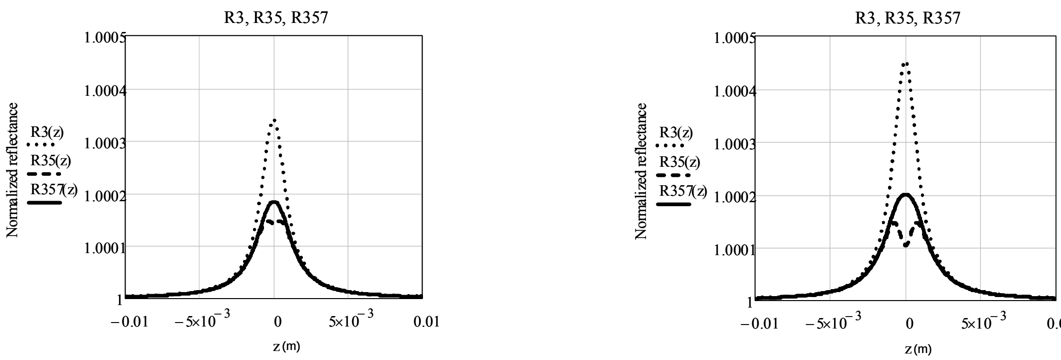
I_0=3 GW/cm^2 I_0=4 GW/cm^2

Fig. 2. The normalized reflectances, R_3, R_5, R_7, if the corresponding nonlinearities of the third-, fifth-, and seventh-order, respectively, would be excited separately.

Fig. 3 shows the normalized reflectances R_3, $R_{3\text{-}5}$, $R_{3\text{-}5\text{-}7}$, in which the contributions of third-order, of third- and fifth-order, and of third-, fifth- and seventh-order nonlinearities, respectively, are considered in the overall nonlinear response of As_2S_3. As already mentioned, due to the opposite signs of n_2 and n_4, in $R_{3\text{-}5}$ curve appears a sharp local valley (the signature of the negative fifth-order nonlinearity) on a larger peak (the signature of the positive third-order nonlinearity).



I_0=1 GW/cm^2 I_0=2 GW/cm^2



I_0=3 GW/cm^2 I_0=4 GW/cm^2

Fig. 3. The normalized reflectances, R_3, $R_{3\text{-}5}$, $R_{3\text{-}5\text{-}7}$, in which are considered the contributions of the third-order, of the third- and fifth-order, and of the third-, fifth- and seventh-order nonlinearities, respectively.

Fig. 4 shows the I_0 dependence of the normalized reflectance peak amplitude for the cases when the third-order, third- and fifth-order, and third-, fifth, and seventh-order nonlinearities, respectively, are considered as contributing to the overall nonlinear response of As_2S_3.

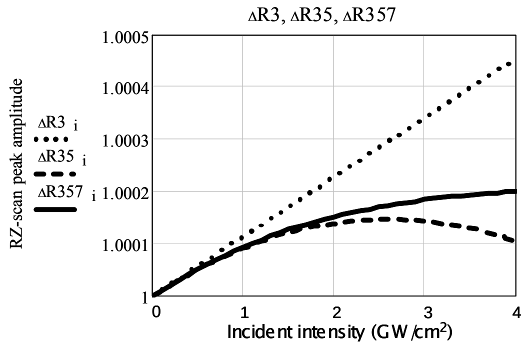
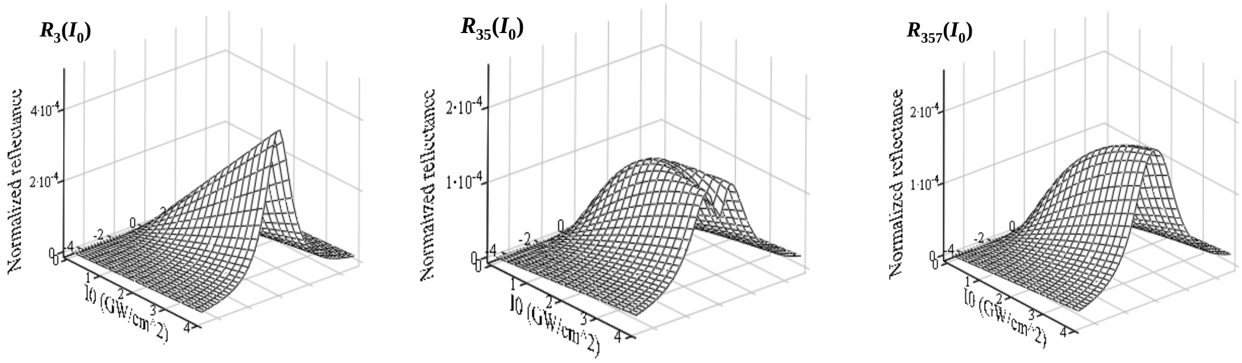


Fig. 4. The dependence of the normalized reflectance peak amplitude on the incident intensity, I_0, for the cases when the third-order (dot line), the third- and fifth-order (dash line), and the third-, fifth-, and seventh-order (continuous line) nonlinearities are considered in the overall nonlinear response.

The linear dependence of the peak amplitude on incident intensity I_0, when only the third-order nonlinearity is considered, as well as the parabolic dependence which appears when the third- and fifth-order nonlinearities are considered, cannot describe the saturating trend of the nonlinear response of As_2S_3. The saturation of the nonlinear response is correctly predicted only when the contribution of the seventh-order nonlinearities are also considered in the overall nonlinear response of the material.

The dependence of the entire RZ-curve on the incident intensity, I_0, for the cases when third-order, third- and fifth-order, and third-, fifth-, and seventh-order nonlinearities are considered in the nonlinear response of the investigated material is shown in Fig. 5.



(a) (b) (c)

Fig. 5. RZ-scan curves as function of incident intensity I_0, when the third-order (a), the third- and fifth-order (b), and the third-, fifth-, and seventh-order (c) nonlinearities are considered in the overall nonlinear response.

The saturating trend of the normalized reflectance of As_2S_3 when the contributions of third-, fifth- and seventh-order nonlinearities are considered together in the nonlinear response of the material is clearly revealed by Fig. 5c.

This prediction of our RZ-scan theoretical model for the dependence of the nonlinear response on incident intensity I_0 is in agreement with the saturating trend of the nonlinearity, experimentally revealed by spectrally resolved two-beam coupling in this material [10].

4. Conclusions

We have presented a theoretical modeling of the normalized reflectance in open aperture reflection Z-scan on nonlinear media with third-, fifth-, and higher-order nonlinearities. A general analytical expression for the normalized reflectance when third-, fifth- and higher-order optical nonlinearities are excited has been derived and its consequences on RZ-scan have been briefly discussed. We have shown that by performing RZ-scan experiments at different incident intensities it is possible to put in evidence the excitation of different order nonlinearities in the medium. Their contributions to the overall nonlinear response can be discriminated by using formulas derived by us. A RZ-scan numerical simulation using our formulas and data taken from literature, measured by spectrally resolved two-beam coupling, for the third-, fifth-, and seventh-order nonlinear refractive indices of As_2S_3 chalcogenide glass, has been performed. Its prediction for the intensity dependent overall nonlinear response of As_2S_3 is in agreement with the one experimentally measured.

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