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Higher order coefficient measurements in nonlinear absorption process.

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Abstract

Intensity-dependent two-photon absorption in chalcogenide glasses has been experimentally observed. Analytical solution of the basic differential equation giving the intensity at the output of the sample is difficult to obtain in this case. A quasi-analytical solution is provided. Second and third-order nonlinear coefficients are deduced from experimental data using Runge-Kutta numerical integration on data obtained via Z-scan technique. Results of the measured higher order nonlinear coefficients are given. Comparison of these results with those obtained by various approximate analytical solutions of the differential equation is made.

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Keywords

Chalcogenide glasses, Nonlinear absorption, Multiphoton absorption, Optical limiting.

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1. Introduction

In recent years chalcogenide glasses have been receiving considerable interest due to their large nonlinearities. Highly nonlinear absorbing materials can find applications in the domain of optical limiting. Ideal optical limiters have large transmittance at low intensities, high pulse energy suppression, a good resistance to laser-induced damage, stability over time and ultrafast responses. Glasses that contain chalcogens (S, Se, Te), are considered as good candidates for optical limiting. Thus, it is of great importance to fully characterize nonlinear absorption properties. In references [1] [2], variation of the two (as well as three)-photon absorption coefficients with the intensity have been reported which suggested the inclusion of higher order effects to better describe the nonlinear absorption.

We have previously investigated nonlinear absorption in some chalcogenide glasses [3,4]. However, in these references, higher order nonlinear absorption effects were not considered. In reference [5], three-, four- and five-photon absorption coefficients are deduced in some chalcogenide glasses from open-aperture z-scan experiments. The coefficients are obtained from a fit of the experimental scan taking into account only one nonlinear absorption process. Recently, in reference [6] we presented a study of nonlinear absorption occurring in telluride based chalcogenide glasses. We observed experimentally an effective two-photon absorption (TPA) process as a function of the incident intensity in four chalcogenide glasses containing different amount of tellurium ($\text{Ge}_{10}\text{As}_{10}\text{Se}_{80-x}\text{Te}_x$ where $x = 0, 10, 15, 20$). We have estimated the values of the respective nonlinear coefficients related to this effective nonlinear absorption. These values were extracted from the experimental data with the help of an approximate theoretical solution of the basic differential equation governing the evolution of the intensity while propagating inside the sample. This solution takes into account the local intensity inside the medium in presence of two-photon absorption. But we have found problem of divergence with the solutions given by this approach.

In this paper, we focus on the exact solution of the basic differential equation relating the intensity inside the medium to the higher order nonlinear absorption coefficients (numerical as well as analytical one). We give the analytical condition necessary to avoid divergence problems. Experimental data are obtained using Z-scan technique [7]. Measurement of the nonlinear coefficient is done by fitting these experimental data with the exact solution given by Runge-Kutta numerical integration method. Comparison of the results is done with those obtained with different simplified solutions. We show that, even though the order of magnitude is respected, the error of the estimated values can be large.

2. Theory

Consider a sample exhibiting the following i) α , the linear absorption coefficient (cm^{-1}) ii) β , the two-photon absorption coefficient (cm/GW) and iii) γ , the third order nonlinear coefficient (cm^3/GW^2). The optical intensity I (in GW/cm^2) satisfies the equation [8]:

$$\frac{dI}{dz} = -\alpha I - \beta I^2 - \gamma I^3 \quad (1)$$

where z is the axis of propagation of light denoting in this section the distance inside the sample. We denote by γ the nonlinear coefficient giving account for the dependency of the TPA coefficient with regard to the intensity I . This coefficient γ denotes here simply a term in the expansion of the absorption in a power series of the intensity that better describes the experimental evolution of the intensity at the output of the specimen (see Refs. [1, 2, 6, 9]).

In the case of a pure TPA phenomenon, the transmission of the intensity through a sample of thickness L (considered as ‘thin’) can be written [10]:

$$T = \frac{I(L)}{I_0} = \frac{e^{-\alpha L}}{1 + I_0 \beta L_{\text{eff}}} \quad (2)$$

where $L_{\text{eff}} = (1 - e^{-\alpha L})/\alpha$, and I_0 is the peak incident intensity at the focus.

When γ cannot be neglected anymore, an effective TPA coefficient β_{eff} has to be introduced. Indeed, it is this value that is usually measured in experiment. An approximate solution of Eq. (1) for the intensity has been provided in reference [6]:

$$I(z) = \frac{I_0 e^{-\alpha z}}{1 + I_0 \left(\beta - \frac{\gamma \alpha}{\beta} \right) L_1(z) + \frac{\gamma (\alpha + \beta I_0)}{\beta^2} \text{Log}[1 + \beta I_0 L_1(z)]}, \quad (3)$$

where $L_1(z) = (1 - e^{-\alpha z})/\alpha$. Comparing relations (2) and (3) for $z = L$, we can easily find:

$$\beta_{\text{eff}} = \beta - \frac{\gamma \alpha}{\beta} + \frac{\gamma (\alpha + \beta I_0)}{\beta^2 I_0 L_{\text{eff}}} \text{Log}[1 + \beta I_0 L_{\text{eff}}] \quad (4)$$

In references [1] and [2], the authors have a more simplified approach to deduce the nonlinear coefficients β and γ . In the case of very thin samples, they write:

$$\beta_{\text{eff}} = \beta + \gamma I_0 \quad (5)$$

We can easily check that when $\beta I_0 L_{\text{eff}} \ll 1$, Eq. (4) reduces to relation (5). Thus, considering the sample as ‘physically very thin’ is not always sufficient to allow the use the simplified expression (5). It depends also on the incident intensity as well as on β , the TPA coefficient. In fact the exact solution for β_{eff} is given by the following (see Eq. (A6) in the appendix):

$$\beta_{\text{eff}} = \beta + \frac{\gamma}{L_{\text{eff}}} \int_0^L I(z) e^{-\alpha z} dz \quad (6)$$

We can see from Eq. (6) that β_{eff} varies as the intensity propagates inside the sample especially in presence of a large TPA coefficient as it can be noticed from the exact solution of Eq. (1) detailed in the appendix:

$$I(z) = \frac{I_0 e^{-\alpha z}}{1 + \beta I_0 L_1 + \gamma I_0^2 L_2^{(\infty)}}. \quad (7)$$

where $L_2^{(\infty)}(\alpha, \beta, \gamma, I_0, z)$ can be easily computed (*cf.* Eq. (A7)). In Fig. 1 is shown a numerical simulation of the intensity evolution while propagating inside the sample for large value of β . The considered parameters are the same as those found in the experiment done with $\text{Ge}_{10}\text{As}_{10}\text{Se}_{60}\text{Te}_{20}$: $\alpha = 3.18 \text{ cm}^{-1}$; $\beta = 13.7 \text{ cm/GW}$, $\gamma = -11.0 \text{ cm}^3/\text{GW}^2$; $L = 0.134 \text{ cm}$; $I_0 = 1.3 \text{ GW/cm}^2$. The quasi-analytical intensity given by Eq. (7) provides the same results when compared to those obtained by using the Runge-Kutta numerical method. These results are significantly different from those deduced from the rough approximations given in Eqs. (3), (4), and (5). In section 3 here below, by comparing the measured values of β and γ using Eq. (3), Eq. (5) and the exact solution, we will illustrate the error induced by these approximations.

3. Experimental results

The glass preparation is described in reference [6]. For optical characterizations, one sample of 16 mm diameter is cut around 1 mm thickness. The sample is polished up to a powder size of 0.5 μm . The optical transparency extends from around 1 μm to 18 μm . Depending on the composition of tellurium in these glasses, we will denote them hereafter, as Te0, Te10, Te15 and Te20. The Z-scan setup is shown in Fig. 2. Excitation is provided by a Nd:YAG laser (Continuum) delivering 15 ps single pulses at $\lambda = 1.064 \mu\text{m}$ with 10 Hz repetition rate. The input intensity I_0 can be varied by means of a half-wave plate and a Glan prism (not shown in this figure), in order to maintain linear polarization. In this work I_0 will be fixed at approximately 1.3 GW/cm^2 , lower than the intensity limit value (I_{lim} in the worst case corresponding to Te20 is equal to 1.35 GW/cm^2), given by the analytical condition (Eq. A3) in order to avoid divergence problem when resolving numerically Eq. (1). Physically, this condition traduces the fact that there is no light emission or amplification inside the sample for large negative value of γ . A beam splitter at the input of the setup

permits to monitor fluctuations occurring in the incident laser beam. Here we assume that the temporal pulse duration is constant and is always equal to 15 ps. Other experimental parameters are: $f = 20$ cm, (focal length of lens L); $d = 26$ cm (distance from the focus to the camera). The beam waist at the focal plane of lens L is $\omega_0 = 35$ μm giving a Rayleigh range $z_0 = \pi \omega_0^2 / \lambda \approx 3.6$ mm. This value is larger than the sample thickness (typically 1 mm). The sample is mounted on a translation stage and moves around the focus region parallel to the beam propagation z-axis. Note that, for Zscan experiment, the origin of this axis is located at the focus of lens L while it was located at the entry of the specimen when considering Eq. (1) in section 2 and in the appendix. The photoreceptor is a 1000×1018 pixels cooled camera (Hamamatsu C4880) with a fixed linear gain. The camera pixels have 4095 gray levels and each pixel is 12×12 μm^2 . Neutral filters ‘nf’ are used to keep the camera within its linear response range. Two sets of acquisitions are carried out for the measurement in order to correct for inhomogenities and surface imperfections in the sample: (i) the low irradiance linear scan is performed by placing the high density neutral filters (nf) before the nonlinear medium (NL). (ii) the high irradiance acquisitions are carried out by placing the same (nf) after the nonlinear material. The latter was subtracted from the first one after normalization as it is recommended in Ref. [7]. No damage or photo-induced phenomena occurred in the tested samples because several experiments have been done showing a good reproducibility of data on the same impact point.

In order to take into account only the contributions of the absorption processes the measurement should be realized after spatially integrating the intensity profiles. We perform the measurement of the nonlinear absorption characterized by the coefficient β_{eff} using the relation for normalized power transmittance [7]:

$$T_p(z) = e^{-\alpha L} \frac{\ln(1 + q_0(z))}{q_0(z)}, \quad (8)$$

where, $q_0(z) = \beta_{\text{eff}} L_{\text{eff}} I_0 / \left[1 + z^2 / z_0^2 \right]$, and I_0 within the sample just after the Fresnel reflection. Eq. (8) gives the ratio of the total transmitted power to that of the incident one obtained by integrating over the radial and azimuthal coordinates for Gaussian incident intensity.

The experiment uses pulses in which the intensity (and hence nonlinear absorption) varies during the pulse. We did not take into account the temporal dependence in this study. A first order correction could be given for a temporally Gaussian pulse. Eq. (8) can be time-integrated to give the normalized energy transmittance. This way of correction is valid only if the response time of detector is much lower compared to the pulse duration. In such a case, we can perform the time-integration because the ‘detection-system’ is considered as linear. In our case, using 15 ps laser pulse, this integration is no more valid. Anyway, a numerical study taking into account a time integration on Gaussian temporal pulses gives a correction factor that shifted our absolute values (given hereafter) without affecting the qualitative evolution of the curves.

The inset of Fig. 3 shows the normalized open aperture Z-scan transmittance for one composition of ChG glass ($\text{Ge}_{10}\text{As}_{10}\text{Se}_{60}\text{Te}_{20}$). In Fig. 3, the transmittance is plotted as a function of $I(z, r=0)$, the on-axis (peak) intensity. This is calculated using $I(z) = I_0 w_0^2 / w^2(z)$, where $w^2(z) = w_0^2 \left(1 + z^2 / z_0^2 \right)$ is the beam radius. The dotted line is the theoretical fit using relation (8). This allows to find β_{eff} , the mean value of the effective TPA coefficient over all the incident intensity range (for $I(z)$ varying from approximately 0 up to 1.4 GW/cm^2). Note that for low intensities the experimental data lie below the best theoretical fit using TPA. On the contrary, for higher intensities the data are above. Similar results are obtained with the other samples with different tellurium concentrations. The corresponding values of β_{eff} are reported in column 6 of Table 1. One can note an increase of this value with tellurium concentration. The solid line in Fig. 3 fits the experimental data more appropriately

because it takes into account the higher order nonlinearity γ . This fitting was done using the function ‘fminsearch’ in Matlab which is based on the Nelder-Mead simplex (direct search) method. For each couple of trial values (β, γ) we used the function ‘ode45’ (based on Runge-Kutta integration method) of the same software in order to resolve numerically the differential equation (1) taking into account Gaussian intensity profiles at the entry and the output of the specimen. At each step we calculated the normalized power transmittance by integrating over the radial coordinate. In columns 7 and 8 of Table 1, we report the measured values of (β, γ) for the different compositions. Table 1 gives also the linear parameters for the four different concentrations of tellurium based chalcogenide glasses. The linear characterization were done using the method described in reference [11]. It can be clearly noted that there is an increase in α , the linear absorption and in n , the linear index, with increasing the tellurium concentration. The increase of α is related to the fact that the band-gap energy of the glass shifts towards lower values when the concentration of tellurium in the glass increases, showing an increase of the semi-conducting character of the compositions. In column 5 we report the normalized photon energy $h\nu/E_g$ with E_g the band-gap energy defined here from λ_{gap} , the band gap wavelength for which $\alpha = 10 \text{ cm}^{-1}$.

4. Discussion

In references [1] and [2] negative values for gamma have been reported for PTS at $1.06 \mu\text{m}$ in the picosecond range using the linear relation (5). In reference [6], we have reported β_1 and γ_1 values for ChG glasses of the same composition, using an imaging technique, with higher intensities (I_0 varied from 1 to 4 GW/cm^2). We have obtained negative value of γ using relations (3) and (4). Even though we obtain approximately the same values for β_{eff} (taking into account the difference of the intensity used in both experiments), the inhomogeneity in these glasses, the use of different measurement method combined with the

experimental error do not permit us to compare precisely the values of β and γ . Another problem comes from the fact that the differential equation (1) diverges for $I_0 > I_{lim}$ (see the Appendix) and this condition was not satisfied experimentally in reference [6]. However, the order of magnitude of the γ values are the same as those obtained here. In order to study more precisely the influence of the simplified relations on the measurement, a comparison is made using the same acquisitions at the same I_0 (below I_{lim}) and the same technique (Z-scan). The obtained values are reported in table 2. In the second column of this table we can find the measured value of β_{eff} . The coefficients β and γ obtained by the exact solution of Eq. (1) are given in columns 3 and 4. Using Eq. (3), the approximate solution (β_1, γ_1) described in reference (6) are given in columns 5 and 6. We also give the solutions β_2 and γ_2 obtained from the simplified approach using the linear relation (5). Note that whatever the simplification is, the measured TPA coefficient (β_1 or β_2) is approximately the same (within the experimental error) as the one given from the exact solution (β). For Te0 where the nonlinear absorption is relatively low compared to the other compositions, there is no need to expand the series in Eq. (1) to the third order. The corresponding values are not given because the uncertainty of the measurement was too high and the results does not have any physical significance. On the contrary, for Te10, Te15 and Te20, the obtained γ values are dependent on the used model. Note that the values of γ_1 obtained using relations (3) and (4) are close to those obtained with the exact solution, while γ_2 values obtained using the linear relation (5) always underestimate γ , the third order nonlinear coefficient. This is due to the fact that in presence of TPA the incident intensity at the output of the specimen is overestimated (see Fig. 1).

In Fig. 4 we can see the evolution of β_{eff} , β and γ with the normalized photon energy. We note a quasi-linear dependence of β_{eff} with $h\nu/E_g$ due to an opposite variation of β , the two-photon absorption coefficient and γ , the third-order nonlinear coefficient. Our quasi-

linear behavior is observed above the half band-gap. In the case of a pure TPA, Sheik-Bahae et al. [12] using a two band model along with Kramers-Krönig transformation have shown that semi-conductors obeys to an universal dispersion curve where a change in the sign of n_2 (nonlinear index coefficient) accompanied with a decrease in β values is predicted for incident pump wavelength close to the band gap (*i.e.* $0.7 < hv/E_g < 1$). However, our results does not follow this trend. In these tellerium ChG glasses, where the normalized photon energy is higher than 0.7, the β values increase with hv/E_g and the sign of n_2 remain positive (shown unambiguously by the peak and valley signature of the Z-scan transmittance giving values of about $+20 \times 10^{-18} \text{ m}^2/\text{W}$ depending on the composition [13]). We do not think that this behaviour is in contradiction with the theoretical and the experimental results of reference [12]. Indeed, the higher order nonlinear absorption phenomena shown in this work can no longer be ignored. Further theoretical investigations are needed to fully understand the response of these glasses when the fifth-order nonlinear susceptibility can not be neglected any more.

5. Conclusion

We have investigated the intensity dependent two-photon absorption in chalcogenide glasses both theoretically and experimentally. We have presented a quasi-analytical solution for the basic differential equation giving the intensity at the exit plane of the specimen under study. The analytical condition that should be satisfied by the incident intensity to avoid divergence problem is provided. Second and third-order nonlinear coefficients are deduced from experimental data using nonlinear fitting combined to Runge-Kutta numerical integration method. Comparison of the results of the measured nonlinear coefficients with those obtained by other approximate solutions of the differential equation is made. We have shown that the exact solution of the basic differential equation describing the propagation of

the intensity inside the medium would give more convincing results than the simplified relations adopted up to now.

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Appendix: quasi-analytical solution for intensity I(z)

The differential equation (1) defining the intensity I as a function of the distance z inside the sample can be analytically solved if: i) we are looking for a solution giving z versus I (this kind of solution cannot be used for comparison with the experimental data), ii) either β or γ are equal to zero. In the latter case ($\gamma = 0$ or $\beta = 0$), we get two Bernoulli's differential equations. The solutions are, respectively:

$$I(z) = \frac{I_0 e^{-\alpha z}}{1 + \beta I_0 L_1}, \quad (\text{A1})$$

$$I(z) = \frac{I_0 e^{-\alpha z}}{\sqrt{1 + 2\gamma I_0^2 L_2}}, \quad (\text{A2})$$

where the functions $L_k(\alpha, z)$ are defined by $L_k = [1 - \exp(-k\alpha z)] / (k\alpha)$ and I_0 stands for the intensity at $z=0$. Note that $L_1(\alpha, L) = L_{\text{eff}}$.

Nevertheless, it is not so easy to find an analytical solution if both β and γ are different from zero. First, in order to describe an absorption process, the derivative dI/dz must be negative. This is true when the coefficients α , β and γ are positive but can be wrong if $\gamma < 0$. In the latter case, dI/dz is negative only if the incident intensity I_0 satisfies the condition:

$$0 \leq I_0 < I_{\text{lim}} = \frac{\beta}{2|\gamma|} \left(1 + \sqrt{1 + \frac{2\alpha|\gamma|}{\beta^2}} \right) \quad (\text{A3})$$

Considering that the condition (A3) is checked or that γ is positive, we can build a trial function compatible with Eqs. (A1-A2) at the $\beta=0$ or $\gamma=0$ limits:

$$I(z) = \frac{I_0 e^{-\alpha z}}{1 + \beta I_0 L_1 + \gamma I_0^2 L_2^{(\infty)}}, \quad (\text{A4})$$

where the function $L_2^{(\infty)}(\alpha, \beta, \gamma, I_0, z)$ necessarily satisfy $\lim_{\gamma I_0^2 \rightarrow 0} L_2^{(\infty)} = L_2$ when $\beta=0$.

Indeed, the latter condition and the differential equation (1) are satisfied if:

$$L_2^{(\infty)} = \int_0^L \frac{e^{-2\alpha z} dz}{1 + \beta I_0 L_1 + \gamma I_0^2 L_2^{(\infty)}}, \quad (\text{A5})$$

Therefore, the intensity $I(z)$ may be numerically computed step by step by using the integral expression:

$$I(z) = \frac{I_0 e^{-\alpha z}}{1 + \beta I_0 L_1(z) + \gamma I_0 \int_0^z I(\zeta) e^{-\alpha \zeta} d\zeta} \quad (\text{A6})$$

Moreover, an approximate value $L_2^{(n)} \approx L_2^{(\infty)}$ can be easily found for each z at any order n .

Considering at the zeroth-order approximation that $L_2^{(0)} = L_2$, we can define this n th-order approximation as:

$$L_2^{(n)}(z) = \int_0^z \frac{e^{-2\alpha \zeta} d\zeta}{1 + \beta I_0 L_1(\zeta) + \gamma I_0^2 L_2^{(n-1)}(\zeta)} \quad (\text{A7})$$

Except in the close neighbourhood of the I_0 limit defined by (A3) in the $\gamma < 0$ case, $L_2^{(n)}(z)$ converges rather quickly (at $n \approx 2$ to 5) towards a stable function and therefore towards $L_2^{(\infty)}(z)$. Putting the latter function in Eq. (A4), we thus get the exact solution $I(z)$ of Eq. (1).

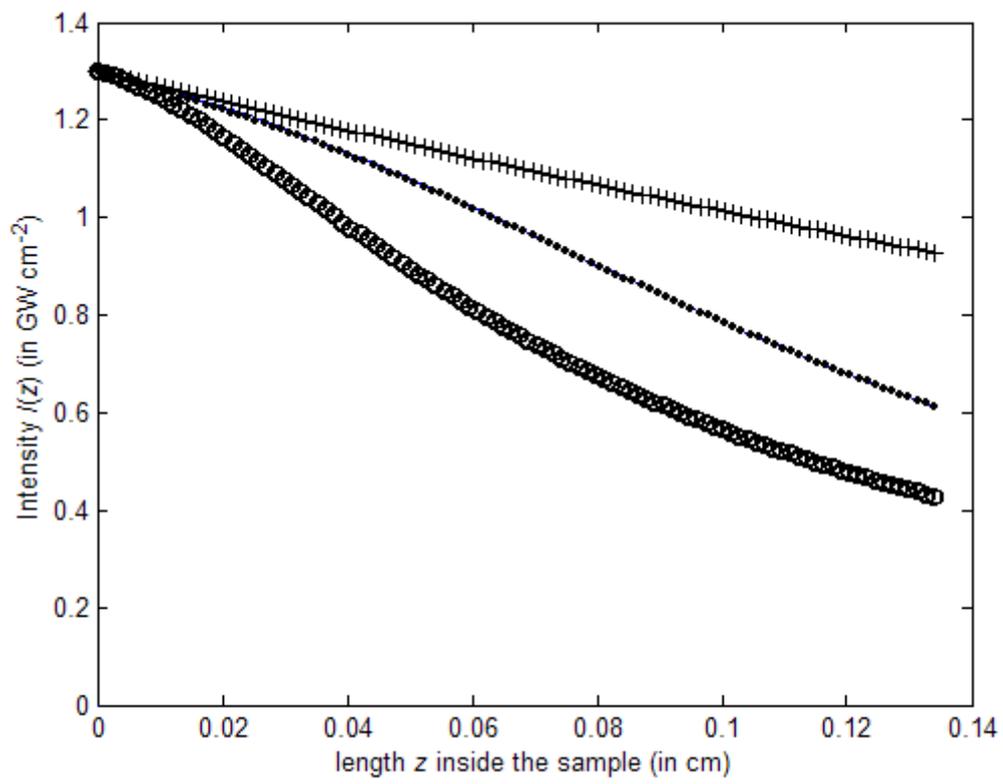


Fig. 1

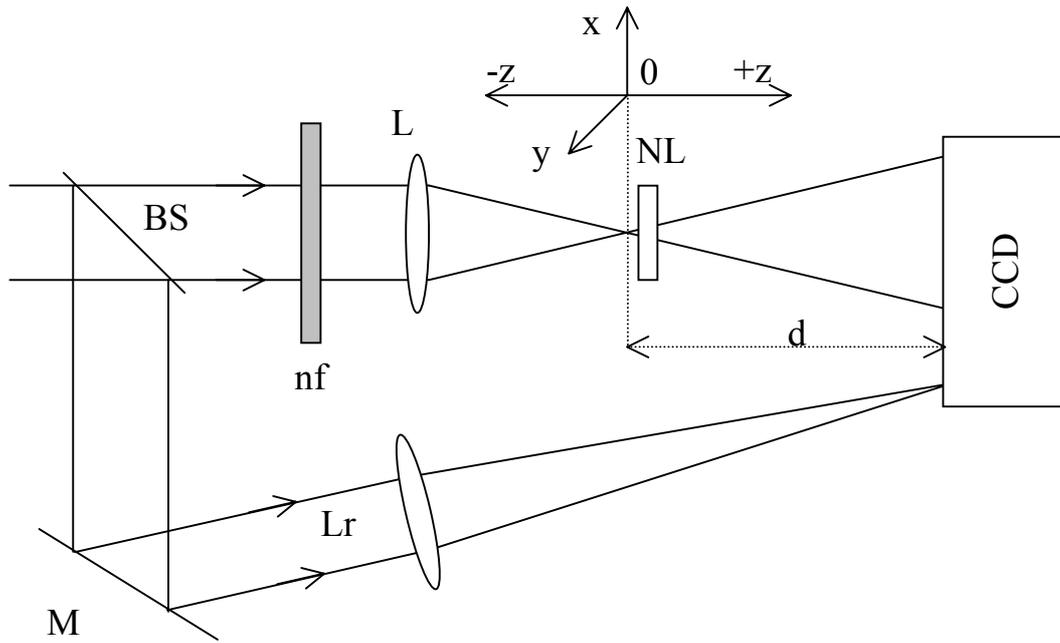
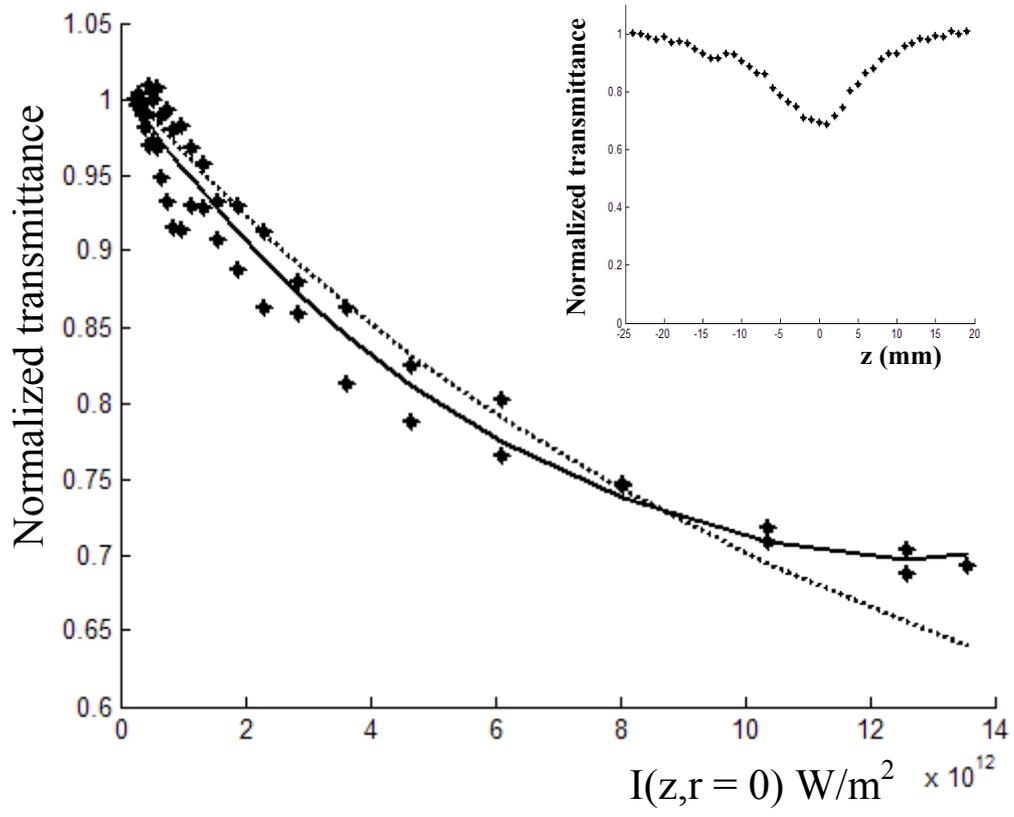


Fig. 2

Figure 3



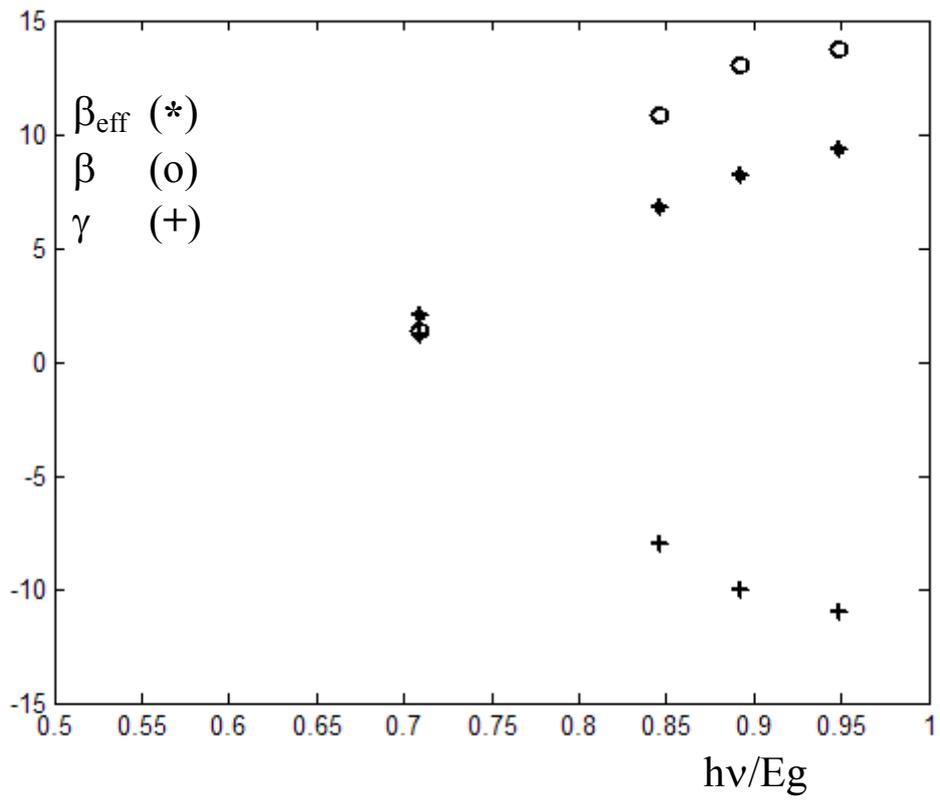


Fig. 4

Table 1

compositions	n	α (m^{-1})	λ_{gap} (nm) ($\alpha = 10 \text{ cm}^{-1}$)	$h\nu/E_g$	$\langle\beta_{\text{eff}}\rangle$ (cm/GW)	β (cm/GW)	γ (cm^3/GW^2)
$\text{Ge}_{10}\text{As}_{10}\text{Se}_{80}$	2.56	14	755	0.71	2.1	1.4	1.2
$\text{Ge}_{10}\text{As}_{10}\text{Se}_{70}\text{Te}_{10}$	2.62	32	900	0.85	6.8	10.8	-8
$\text{Ge}_{10}\text{As}_{10}\text{Se}_{65}\text{Te}_{15}$	2.75	72	950	0.89	8.2	13	-10
$\text{Ge}_{10}\text{As}_{10}\text{Se}_{60}\text{Te}_{20}$	2.80	318	1010	0.95	9.3	13.7	-11

Table 2

compositions	β_{eff}	β	γ	β_1	γ_1	β_2	γ_2
Te0	2.1	-	-	-	-	-	-
Te10	6.8	10.8	-8	11.2	-10	10.5	-4.8
Te15	8.2	13	-10	11.7	-8	11.2	-4.3
Te20	9.3	13.7	-11	14.6	-14	13.6	-5.9

Figure captions

Fig.1

Numerical simulations of the intensity evolution while propagating inside a sample for $\text{Ge}_{10}\text{As}_{10}\text{Se}_{60}\text{Te}_{20}$ with the parameters: $\alpha = 3.18 \text{ cm}^{-1}$; $\beta = 13.7 \text{ cm/GW}$, $\gamma = -11.0 \text{ cm}^3/\text{GW}^2$; $L = 0.134 \text{ cm}$; $I_0 = 1.3 \text{ GW/cm}^2$. (+): approximation defined by Eq. (5); (o) approximation given by Eqs. (3) and (4); (●) exact solution of Eq.(1).

Fig.2

Experimental setup (Z-scan). M: mirror; BS: beam splitter; L, Lr: lenses, nf: neutral filters, NL: nonlinear material. 'd' is the distance of the CCD camera from the focus.

Fig. 3:

Transmission T_p versus the on axis peak intensity $I(z)$ for $\text{Ge}_{10}\text{As}_{10}\text{Se}_{60}\text{Te}_{20}$. The stars represent the experimental data points; the solid line is the best theoretical fit giving $\beta_{\text{eff}} = 9.3 \text{ cm/GW}$. In the inset is shown the experimental open aperture normalized Z-scan transmittance (stars) versus z , the position of the specimen.

Fig.4

Evolution of β_{eff} , β , and γ with the normalized photon energy $h\nu/E_g$.

Table Captions

Table 1.

Linear and nonlinear optical coefficients at 1.06 μm for different chalcogenide glasses in the picosecond range.

Table 2.

Comparison of higher order nonlinear absorption coefficients obtained from various approximations for different compositions. In the column 2 is given β_{eff} , β and γ are the values given by the Runge-Kutta numerical integration (exact solution). β_1 , γ_1 are those obtained from Eq. (3). β_2 , γ_2 are deduced from the more simplified linear relation (5). The β values are given in cm/GW and γ in cm^3/GW^2 .

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