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IMPROVEMENTS OF THE Z-SCAN METHOD USED IN THE STUDY OF OPTICAL NONLINEARITIES

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Z-scan method used for the study of optical nonlinearities in materials has been improved on the basis of a complex analysis of the laser beam. Using a CCD camera for beam intensity distribution measurement and a soft aperture, there was demonstrated the possibility to rise significantly the sensitivity of the method and the signal to noise ratio of the Z-scan curves. An improved two-dimensional measurement procedure for induced lens is proposed. The étalon effect and the influence of the high nonlinear absorption on the transmission of silicon samples were investigated in the case of laser pulses of microsecond and nanosecond length.

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1. Introduction

The principles of the laser based optical methods in the study of materials, consist in the measurement of the modification of laser beam parameters (intensity, direction of propagation, polarization, etc.) as a result of the light-sample interaction. A lens effect induced during propagation of a gaussian laser beam through an optical nonlinear sample will change the parameters of the beam in a predictable manner, in the first order approximation [1]. By measuring the variation of the beam parameters it is possible to deduce the parameters of the lens induced into the material.

A wave travelling in a medium suffers phase changes induced by the non-linear modification of the refractive index. These changes can be detected by several beam-distortion methods. In some cases these methods allow to measure the absorption changes, too.

The optical nonlinearities producing these effects have various origins [2-12]. In resonant conditions, when absorption is present, the predominant effect is a thermal change of complex refractive index (real and imaginary part). Superimposed to this contribution there are effects connected to resonant excitation of carriers or nonresonant distortion of electronic orbits. Non-resonant third-order nonlinearities have very short response times but usually are weak [6,7,13,14].

The Z-scan or self-diffraction method is one the most useful method for the measurements of the optical nonlinearities, based on the induced lens effect. In particular, nonlinear refraction and nonlinear absorption can be followed by this technique [14].

The use of the Z-scan method is limited by the noise introduced due to non-uniformity of the intensity distribution and fluctuations of the laser beam (intensity, optical axis of the beam) used in experiments.

This paper shows the results obtained by us in the improvement of the Z-scan standard procedure, starting from a careful analysis of the characteristics of the laser beam and of the geometrical conditions specific to standard Z-scan procedure. A simple, two-dimensional method, based on the analysis of the probe beam parameters was developed and the experimental results are discussed for a doped silicon sample.

2. Z-scan method

In this method, the transmission of a focused laser beam through a finite aperture in the far field is measured as a function of the displacement (z) of an optically nonlinear sample along the propagation direction with respect to the focal plane [14] (Fig. 1). The transmission curve obtained can be understood by considering the sample as a thin lens whose focal length is determined by the nonlinear change of the refractive index produced by the intensity on the sample itself.

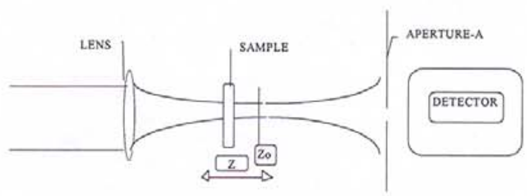


Fig. 1 Z-scan experimental set-up.

When a laser beam propagates through a medium with nonlinear response, the radial distribution of the beam intensity induces a distribution of the refractive index (or/and absorption coefficient) which in turn modifies the propagation of the beam itself. This modification of the refractive index acts as a positive or negative lens according to the sign of the nonlinear coefficient. A fast variation of the beam intensity distribution, which means a fast variation of the induced changes, is obtained by using a focused beam. The basic idea of the self-diffraction method, in its standard variant, is to scan the sample through a focused beam and to measure the profile of the transmitted signal through an aperture positioned at a fixed distance from the waist of the focused beam. The profile and the amplitude of this transmission contains all the necessary information for the determination of the sign and value of the nonlinear parameters.

The effect of the refractive index distribution on the beam propagation has been studied firstly, in the frame of the theoretical model proposed by Sheik-Bahae et al. [14] which uses the Slowly Varying Envelope Approximation (SVEA) developed by Weaire et al. [15]. The effect of refractive index distribution was discovered several years before [16].

The model is valid for thin samples as compared to Rayleigh length of the incident beam. The model was extended to thick samples [17-19], non-gaussian beams and étalon effect [18-22]. For thick samples two rigorous methods were developed: the analytical treatment developed by Hermann and McDuff [21], corrected to first order in irradiance and the numerical Gaussian-Laguerre modal decomposition developed by McDuff [21,22]. In all the models the sensitivity of the nonlinear parameters is given by the sensitivity in the measuring of the optical transmittance for which the aperture size and position are important parameters.

In the standard variant, for the case of a sample with Kerr nonlinearity, the dependence of the normalized transmission, T, through the aperture, as a function of the sample position, z, is defined using transmitted and incident power, P_t and P_i, by the equation [14]:

$$T(z) = \frac{\int_{-\infty}^{\infty} P_t(\Delta\Phi_0(t))dt}{S\int_{-\infty}^{\infty} P_i(t)dt} \tag{1}$$

S is $1-exp(-2r_a^2/w_a^2)$, where r_a is the aperture radius and w_a is the beam radius at the aperture position in the absence of nonlinearity.

The transmission exhibits several interesting features:

- a peak-valley evolution for a negative nonlinearity and a valley-peak for positive one;

- the separation between the minimum and the maximum remains nearly constant and is proportional to Rayleigh length;

- the phase shift is proportional to the maximum variation of the transmittance ΔT_{p-v}; a smaller value of the aperture radius produces a larger value of ΔT_{p-v} that means a higher sensitivity.

The model is valid only for very thin samples, if compared to the Rayleigh range of the incident beam. The other models try to extend the theory to thick samples, non-gaussian beams or higher orders in irradiance dependence but they use the same procedure. We will analyse the possibility to optimize the parameters of the aperture taking into account the noise sources. Most of the noise, due to optical linear effects (sample inhomogeneities, quality of the optical surface of the sample, etc.) can be removed with the help of a Z-scan procedure at low intensities. The pointing stability of the laser beam was, in general, fully ignored, although this is an important source of noise, as we will see. In order to make corrections for this effect and to improve the sensitivity of the transmission measurement we used a "soft" aperture technique: the laser beam intensity distribution is registrated at the aperture position using a laser beam analyser with a CCD camera. The obtained distribution is analysed with a numerically simulated aperture.

3. Results and discussion

3.1 Aperture optimization

As already shown, the essential fact in the standard method is the accurate measurement of the normalized transmittance. We studied the influence of the pointing stability of the laser beam on the transmitted signal through the aperture A, using various aperture sizes. The results for three sizes are presented in Fig. 2 for the range of typical beam pointing stability of usual laser systems. The apertures of various sizes exhibit various sensitivities to the changes of the beam axis. An increase of the noise effect is obtained when the diameter of the aperture decreases.

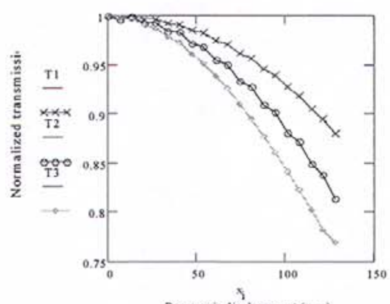


Fig. 2 The normalized transmissions T1, T2, T3 through the apertures of diameters 1.2w, w and 0.8w.

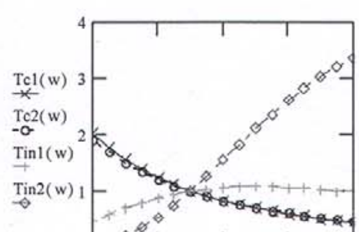


Fig. 3 Modification of the signal intensity transmitted (a.u.) through circular aperture: Tc1and Tc2, corresponding to aperture radius of $0.1w_a$ and $0.5w_a$, respectively) and ring apertures ($r_I=1.1w_a$, $r_{ex}=1.2w_a$; $r_I=1.4w_a$, $r_{ex}=1.5w_a$) as a function of the increase in the beam diameter at the aperture position.

Fig. 3 shows that the variation of the signal transmitted through the aperture is larger when a ring type aperture is used, with the lowest radius greater than the beam diameter, in the absence of the nonlinear effect, w_a.

To improve the Z-scan sensitivity and to reduce the noise on the Z-scan curve a "soft" aperture is the best choice. The beam intensity distribution is registered with a laser beam analyser and a CCD camera with good resolution. The size and position of the aperture can be optimized by numerical methods. To minimize the influence of the pointing stability of the beam the centre of the "soft" aperture is positioned each time at the centroid position of the intensity distribution. The centroid position is not modified by the symmetric changes of the intensity distribution due to induced lens. For a given angular pointing stability of the laser beam, the position of the aperture must to be situated as near as possible to the beam waist position, in order to minimize this influence. Two Z-scan signatures (with and without aperture) must be registered in order to make the separation between the nonlinear refractive index and the nonlinear absorption. When the numerical analysis of the intensity distribution is used, as in our procedure, the two signatures can be obtained simultaneously. The second detector, used in standard experimental configuration for the subtraction of the beam intensity fluctuations, is not necessary. If the incident beam, with a circular symmetry, becomes elliptical, the ellipticity is a measure for the anisotropy of the nonlinear property in the sample. A two-dimensional analysis can be done from the elliptical dimensions of the beam, using two perpendicular diameters.

3.2 Two-dimensional analysis of the nonlinear parameters

The determination of the optical nonlinear coefficients by the Z-scan method is based on the measurement and analysis of the spatial beam-profile distortion caused by the optical nonlinearity of the sample. Firstly, this means that the spatial profile of the beam must be accurately characterized and controlled. The laser beam propagation can be described if the following parameters are determined [1,23,24]: the beam waist position, z_o; the beam waist diameter, w_o; the angle of divergence in far field, θ. The laser beam propagation has some invariance properties described by the equation:

$$n d_{\sigma o} \theta_\sigma = \frac{M^2 4 \lambda_o}{\pi} \tag{2}$$

an equivalent formulation of the Lagrange-Helmholtz invariant. The significance of the symbols are those well known: n the refractive index of the propagation media, λ_o, θ_σ – the wavelength of the beam and the divergence, respectively; $d_{\sigma o}$ is the beam diameter. M^2 parameter shows how many times the real beam divergence is larger than that of a gaussian beam with the same parameters. The divergence can be written as a function of the beam diameter and the diameter becomes the most important parameter for the laser beam change analysis during its propagation. The beam diameter may be defined in a number of ways, but the definition using the second order moment σ of the intensity distribution of the beam reflect more deeply the physical meaning of this parameter. For beams with ellipticity, two perpendicular diameters will be defined in a same manner.

Any definition is used for the beam diameter, the most important fact is that the ratio of the effective beam diameter $d(z)$ to the underlying gaussian spot size $w(z)$ will remain constant at all planes z, independent of the actual multimode beam profile [1,24]. The equation of propagation of the gaussian beam diameter $w(z)$ is still valid for the multimode beam diameter $d(z)$:

$$d^2(z) = d_0^2 + \left(\frac{M^2 \lambda}{\pi d_0}\right)^2 (z - z_0)^2 \tag{3}$$

The invariance properties of the laser beam propagation are conserved and after the propagation through optical systems. We propose, according to the invariance properties, to use the parameters of the spatial profile of the beam, essentially the beam diameter, to calculate the values of the nonlinear parameters. The analysis will be carried out for the standard procedure case: for thin sample and for gaussian beam. Using the previous considerations [1,19,24], it can be easily extended to elliptical non-gaussian beam, in order to develop a two-dimensional analysis.

For a thin sample, the effect of inducing a profile for the refraction index is equivalent to the formation of a thin lens. For thick samples one applies the matrix decomposition procedure for complex optical systems [1,17]. The beam diameter at the aperture position, after the propagation through a thin lens with the focal length f(z,γ) dependent on its position z and the nonlinear coefficient γ of the sample, can be written:

$$d'^2(D,z,\gamma)=\frac{f^2(z,\gamma)d_0^2}{\left(z-f(z,\gamma)\right)^2+z_R^2}\left[1+\frac{\left[\left(z-f(z,\gamma)\right)^2+z_R^2\right]^2}{z_R^2 f^4}\left(z-D+f(z,\gamma)+\frac{f^2(z,\gamma)\left(z-f(z,\gamma)\right)}{\left(z-f(z,\gamma)\right)^2+z_R^2}\right)^2\right] \tag{4}$$

$f(z, \gamma)$ is the induced equivalent focal length, d_o and z_R are the waist diameter and Rayleigh length of the focused beam, D is the position of the aperture referred to the waist of the focused beam position; g and γ are the thickness and the nonlinear reffractive index of the sample, respectively. The dependence on intensity of the induced lens is obtained using different assumptions regarding the radial profile of the nonlinear refractive index. For a sample of thickness g in the parabolic approximation, the refractive index is:

$$n(r) = n_0(1 - ½\, \delta n{\cdot}r^2) \tag{5}$$

The equivalent focal length, in thin lens approximation, is:

$$f \approx (\delta n{\cdot}g)^{-1} \tag{6}$$

For a gaussian beam profile, with diameter in the sample w, which induces a maximum refractive change Δn_0 on the axis a similar relation to (4) can be used:

$$n \approx n_0(1 - 2\Delta n_0{\cdot}r^2/n_0{\cdot}w^2) \tag{7}$$

The approximated focal length is:

$$f(z)=\frac{an_0\left[w(z)\right]^2}{4\Delta n_0(z)d} \tag{8}$$

where a is a constant which can be introduced for the correction of the difference between the real gaussian profile and approximated profile [19].

Using the assumption $\Delta n_0(z)=\gamma I_0(z)$, where I_0 is the beam intensity on the axis, at z position, the relation between the power of the beam P and I_0 : $I_0(z)=2P/\pi w^2(z)$, and the equation (3) the induced focal length is written as a function of the beam parameters and sample's nonlinear parameter at any plane z along the direction of propagation:

$$f(z,\gamma)=\frac{\pi n_0 d_0^4}{8\gamma g P}\left[1+\left(\frac{z}{z_R}\right)^2\right]^2 \tag{9}$$

By introducing (9) in (4) one obtains the dependence of the beam diameter on z and on nonlinear parameter γ. By comparing the measured diameter at different positions z with the diameter extracted from (4) the nonlinear parameter, γ, can be calculated.

3.3 The étalon effect

An other important noise source is the so-called étalon effect: due to the high value of the refractive index of the sample, the multiple internal reflections on the sample's faces will change the field intensity within the sample and the sample transmission [1]. This effect can disturb profoundly the Z-scan signature [20]. When the Fresnel reflections and the nonlinear coefficients are small, the effect can be subtracted using the well known procedure [11,18,20]. However for samples with high refractive index a large variation of the nonlinear parameter makes this problem more difficult. In Fig. 4 is presented the transmission of a doped silicon sample (4 Ωcm) obtained in a Z-scan measurement with open aperture, for a Nd:YAG laser. The maximum incident intensity is 2×10^3 W/cm^2. Fig. 5 shows the calculated transmission of a Fabry-Pérot étalon (dispersive Fabry-Pérot étalon) with the same parameters as silicon sample (thickness and reflectivity) and a phase shift variation produced by carrier pairs generation at the same incident intensity as in the experimental case.

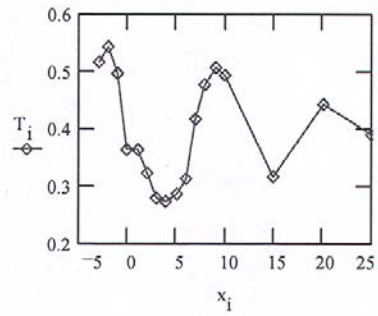


Fig. 4 Experimental Z-scan signature for a doped silicon sample at incident intensities up to 2×10^3 W/cm^2 at the beam waist position and for 22 ns pulse length.

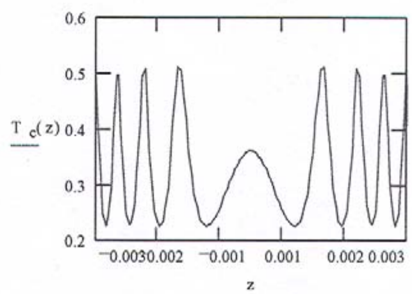
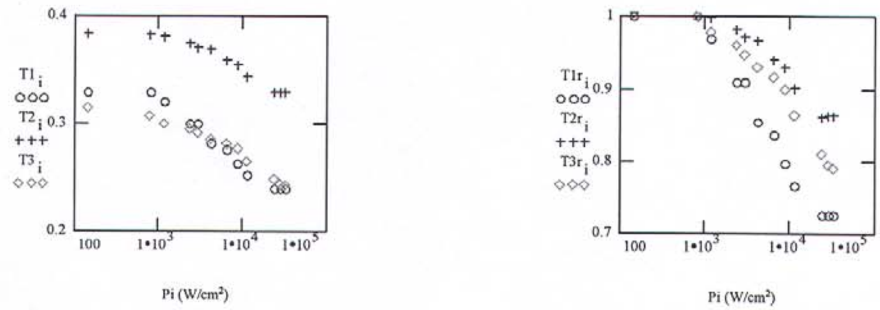


Fig. 5 Transmission of a dispersive Fabry-Pérot étalon with the same parameters as the silicon sample, a phase shift due to pair generation at incident intensities up to 2×10^3 W/cm^2 at the beam waist position and for 22 ns pulse length.

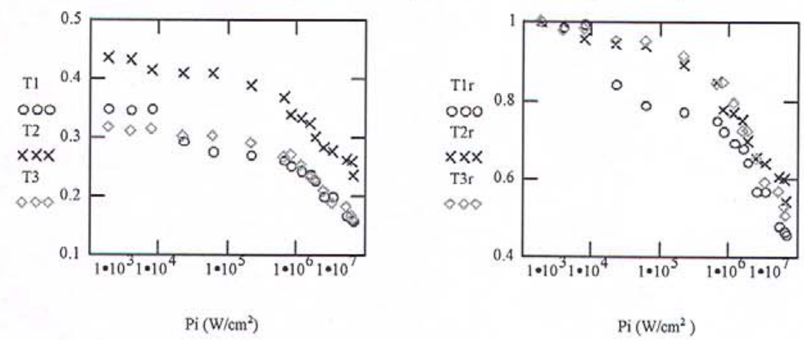
As can be seen, the Z-scan signature near the beam waist is practically determined by the dispersive étalon effect. At higher incident intensities the nonlinear absorption becomes significant and the pure dispersive étalon effect strongly decreases. The influence of the increasing absorption on the étalon transmission becomes predominant. The transmission obtained as a function of incident intensity for three samples and two laser pulse durations are presented in Fig. 6 and 7.



a) b)

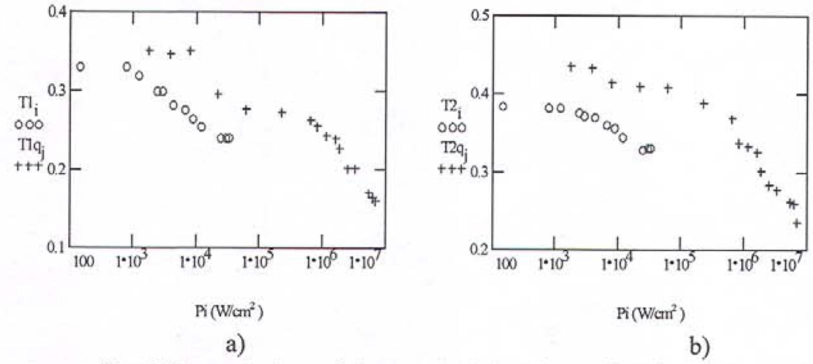
Fig. 6 Transmissions a) and relative transmissions b) for three doped silicon samples (Nd:YAG laser with pulse duration of 50μs).

The main contribution to the nonlinear absorption increase seems to be the increase of the free rier absorption. The reflectivity increase and the two-photon absorption contributions are too small l cannot explain the strong nonlinear coefficient obtained. At the same time the threshold of the ilinear transmission is situated at fluences of the incident beam capable to generate densities of the e carriers higher than those existing in the sample due to the impurities ($>2\times10^{15}$ cm^{-3}).



a) b)

Fig. 7 Transmissions a) and relative transmissions b) of three doped silicon samples T1, T2, T3 (Nd:YAG laser with pulse duration of 22 ns and near gaussian time profile).



a) b)

Fig. 8 Transmissions of the sample 1 a) and sample 2 b) for the 50μs (ooo) and 22 ns (+++) laser pulse durations.

The signature of the étalon effect is not directly observed but it still influences the nonlinear transmission of the sample: its superimposed modulation on the transmission curve seems to be higher for the sample with better parallelism between the faces. This makes almost impossible the separation of different nonlinear contributions. Only an antireflection coating can minimize it. In the presence of the étalon effect, a high nonlinear absorption distorts more deeply the Z-scan signature. The value of the incident intensity must be optimized to keep the nonlinear absorption at low level. This can be made easily by monitoring the shape of the intensity beam distribution: an increasing absorption produces a stronger effect on the higher intensity zone (the beam becomes top-hat) and a saturable absorption makes the distribution sharper than in the absence of the nonlinearity.

3.4 The laser pulse length effect

By comparing the transmissions of the sample at the microsecond and nanosecond laser pulse length, different values were obtained at the same values of the maximum power densities (Fig.8). This can be explained only by the response time of the nonlinearity which is dependent on the main sources of nonlinearity. The estimated rise time of the field in an empty Fabry-Pérot interferometer with our sample's parameter is of the order of 200 ps. This is much lower than laser pulse length and its influence can be considered insignificant. That means the choice of the laser pulse length must correspond to the time response of the nonlinearity to be studied. Generally the rise time of the nonlinearity is much lower than the relaxation time. For our samples, the recombination time of the carriers has the main contribution. The pulse length must be much shorter or much longer than the response time, in order the model developed for the influence of the pulse length [12,14] to be still valid. A pulse length of the order of the time response of the main nonlinearity affects the validity of the model.

4. Conclusions

A careful analysis of the sources of noise and distortion in the Z-scan method allowed for significant improvement of this method used in the investigation of the optical nonlinearities in transparent materials. The optimization of the aperture size and the choice of the optimum position referred to the laser beam waist position, significantly reduce the noise in the transmitted optical signal and minimize the influence of the fluctuations of the laser beam, thus allowing for accurate measurement of the optical nonlinear parameters. The complex analysis of the laser beam allows for a simple two-dimensional measurement of the sample nonlinearity in order to determine the existence of the optical nonlinear anisotropy. The analysis of the experimental results obtained on some crystalline silicon samples has demonstrated that: for doped silicon the free carrier absorption seems to be the main contribution to the increase of nonlinear absorption. It is necessary to use the appropriate length pulses and incident fluences in order to decrease the influences of some additional distortions on the Z-scan signature.

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