



Sensitivity of Z-scan using diffraction efficiency

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ABSTRACT

We report Z-scan and eclipsing Z-scan experimental traces measuring diffraction efficiency in the image plane of the 4f system. Using nonlinearly diffracted energy, the sensitivity of both techniques is compared in order to increase the signal-to-noise ratio. The optimization of the optical signal for nonlinear characterization is proposed. A simple linear relation is provided in order to characterize nonlinear refraction from diffraction efficiency measurements. The influence of the nonlinear absorption is discussed.

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1. Introduction

In the metrology, the sensitivity is a parameter describing an ability of experimental technique for measuring small changes in physical quantities. This factor is particularly important in nonlinear optical characterization where relatively small intensity-dependent modifications in refraction and absorption must be transferred into measurable quantities. Among many nonlinear characterization techniques, the Z-scan method [1,2] was found to be especially efficient in converting the small nonlinear response of the material into a detectable signal. In this method, the sample is scanned along the optical path (z -axis) of a focused Gaussian beam, which undergoes phase and amplitude changes after passing through a nonlinear medium. The resulting beam distortion is amplified in the diffracted far field and then quantified by measuring the energy through a small circular aperture in function of z , the sample position. In usual procedure, the diffracted energy detected in the nonlinear regime ($\varepsilon_{D_{NL}}(z)$) is normalized to the energy obtained in the linear regime ($\varepsilon_{D_L}(z)$) defining a normalized Z-scan transmittance ($T_{\text{nor}}(z) = \varepsilon_{D_{NL}}/\varepsilon_{D_L}$). The latter has a characteristic peak-valley configuration from which the nonlinear refraction is estimated. The sensitivity of this method increases with decreasing aperture size allowing to sense a nonlinear wave front distortion as small as $\lambda/250$. Later, several variations of the Z-scan method have been developed in order to improve this sensitivity [3–9]. Particularly, the need to characterize thin film samples has motivated the development of the eclipsing Z-scan (EZ-scan) where the disk is used instead of the aperture [10] and the transmittance of the beam around this obscuration is

measured. Using the same normalization procedure (T_{nor}) as in the original Z-scan the possibility of measuring a wave front distortion of $\approx \lambda/10^4$ has been estimated for a very large disk (more than 98% of stopped light) and a signal-to-noise ratio of unity. In this article, we will show that this improvement of sensitivity is rather due to the more significant reduction of linear signal (ε_{D_L}) with respect to nonlinear signal than due to the higher nonlinear distortion of the beam in its wings. This problem has been already mentioned in Ref. [11], where the combination of the Z-scan technique with the binary diffractive elements has been proposed. Another inconvenience related with the normalization to the linear regime is that the improvement of the sensitivity in Z-scan (EZ-scan) experiment with the decrease (increase) of the aperture (disk) size always comes at the expense of a reduction in accuracy due to the decrease of signal-to-noise ratio. In the context of these drawbacks, it would be valuable to define normalization procedure where no problem of division by very small signal (division by “zero”) could happen, and consequently, the comparison of optical sensitivities due to a pure nonlinear response of the sample would be possible.

An application of two-dimensional CCD camera in Z-scan measurements [12–14] has provided more flexibility with processing the measured signal since the total far field pattern of diffracted beam can be carefully studied. The acquired data are digitized and stored in a computer allowing a numerical analysis of the recorded images. With these facilities, there is no need to use physical aperture or disk at the output of experimental setup since the numerical ones can be applied. Moreover, the spatial accuracy of measurements is increased because the size of the pixels may be as small as few micrometers and both signal and reference beams can be recorded simultaneously to eliminate laser fluctuations. We have exploited these properties of two-dimensional detection introducing different nonlinear characterization techniques [15–20] inside a 4f coherent imaging system

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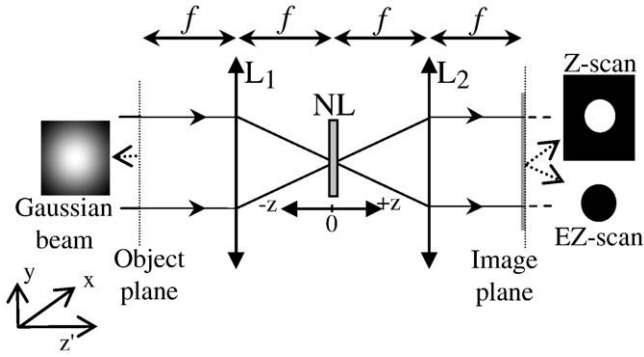


Fig. 1. Schematic of the 4f imaging system. The nonlinear medium (NL) is moved around the focal plane. The labels refer to lenses (L_1 – L_2) and their focal lengths (f). The object at the entry is the Gaussian beam, while a circular aperture or an opaque disc is placed in the image plane to obtain Z-scan or EZ-scan configuration, respectively.

(Fig. 1). Different objects placed at the entry of system and the optimized field stops, situated the image plane, were proposed in order to analyze the image intensity profiles after nonlinear filtering through the material (NL) placed in focal region. For quantitative description of nonlinearly diffracted signal, we have defined diffraction efficiency as the signal to measure [18–20]: $\eta = \varepsilon_{D_{NL}}/\varepsilon_T$, where $\varepsilon_{D_{NL}}$ is the diffracted energy in nonlinear regime and ε_T is the total energy detected in the nonlinear regime without any field stop at the output. The latter quantity is typically two orders of magnitude higher than the diffracted nonlinear signal ($\varepsilon_T \gg \varepsilon_{D_{NL}}$), thus no problem of division by zero could happen. Moreover, we have shown that by choosing an appropriate object at the entry and a field stop in the image plane, we can perform various well-known experiments as Degenerate Four Wave Mixing (DFWM) [21,22], Z-scan, EZ-scan, I-scan [23] or Nonlinear Imaging Technique with a Phase Object (NIT-PO) [15]. Therefore, all these seemingly different methods can be considered as special cases of the same nonlinear imaging process. Consequently, the measurements of nonlinearly diffracted energy in the image plane of a 4f system provide a good tool for comparison of optical sensitivities corresponding to different experimental techniques. One of the particular cases examined numerically in Ref. [19] was the I-scan experiment, i.e., the 4f system with the Gaussian beam at the entry and a circular aperture or an opaque disc at the output (as shown in Fig. 1) for a sample position fixed in the focus. Here we will describe in details the sensitivity of the same system however considering different positions of the sample in the focal region. Hence, in fact, we will investigate the classical Z-scan and EZ-scan experiments with the diffraction efficiency as a signal to be measured. It has been already checked [24] numerically and experimentally that the addition of lens L_2 (see Fig. 1) does not affect the results of Z-scan. In fact, this lens contributes to produce the Fourier transform of the field at the exit surface of the sample, which is physically similar to the far field diffraction pattern obtained with the original Z-scan method. In this article, we will compare experimentally and numerically the sensitivity of Z-scan and EZ-scan experiments due to pure nonlinear response of the sample where no division by small linear signal could happen. Moreover, we will show that by using the diffraction efficiency, the optimization of the sensitivity can be accompanied simultaneously by an enhancement of the signal-to-noise ratio. The conditions for optimizing pure nonlinear refractive signal in Z-scan measurements will be provided. The influence of nonlinear absorption will be also discussed.

2. Theoretical model

For self-consistency, let us briefly recall the general scheme of beam propagation inside the 4f imaging system (see Fig. 1) described in details in Ref. [18]. The amplitude of electric field distribution at the

object plane is Gaussian, $E(x,y) = E_0 \exp[-(x^2 + y^2)/\omega_e^2]$, where x, y are the spatial coordinates, E_0 denotes the on-axis amplitude, and ω_e is the beam waist at the entry of the setup. Let $S(u,v)$ be the spatial spectrum of $E(x,y)$:

$$S(u,v) = \tilde{F}[E(x,y)] = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} E(x,y) \exp[-j2\pi(ux + vy)] dx dy, \quad (1)$$

where $\tilde{F}$ denotes the Fourier transform operation, u and v are the normalized spatial frequencies. The spectrum of the object is propagated over a distance z' , by taking into account the transfer function of the wave propagation phenomenon [see Chapter 3 in Ref.

[25]] $H(u,v) = \exp\left(j2\pi z' \sqrt{1 - (\lambda u)^2 - (\lambda v)^2} / \lambda\right)$, where λ is the wavelength. The field amplitude at z' after the free propagation is obtained by computing the inverse Fourier transform: $E(x,y,z') = \tilde{F}^{-1}[S(u,v)H(u,v)]$. To calculate the output beam after passing through a lens focal length f , we apply the phase transformation related to their thickness variations in paraxial approximation: $t_L(x,y) = \exp[-j\pi(x^2 + y^2)/\lambda f]$. The first propagation is performed on a distance $z' = f$ from the object plane to the lens L_1 . Then we propagate the beam up to the sample located at z using $z' = f + z$ in H the optical transfer function ($z = 0$ at the focus of the lens L_1). After that, the nonlinear response of the material is taken into account. We assume cubic nonlinearity and a thin nonlinear medium of thickness L exhibiting (i) linear absorption defined by α (m^{-1}), (ii) two-photon absorption defined by β (m/W), and (iii) nonlinear refraction defined by n_2 (m^2/W). In these conditions, the transmittance of the sample is described as [2]:

$$T(u,v,z) = e^{-\alpha L/2} \{1 + q(u,v,z)\}^{j\Delta\phi_0/q_0 - 1/2}, \quad (2)$$

where $q(u,v,z) = \beta L_{eff} I(u,v,z)$. Here $q_0 = \beta L_{eff} I_0$ and $\Delta\phi_0 = 2\pi L_{eff} n_2 I_0 / \lambda$ denote the on-axis nonlinear absorption and nonlinear phase shift at the focus, respectively. Whereas $L_{eff} = [1 - \exp(-\alpha L)]/\alpha$ is the effective length and I_0 is the focal on-axis intensity. Next, we perform propagation on a distance between sample and lens L_2 , and the final diffraction is calculated with $z' = f$ up to the image plane. The image intensity ($I_{im}(x,y)$) at the output is obtained by squaring the amplitude field. The total energy in the output plane is achieved by integrating spatially over the entire image: $\varepsilon_T = 2\pi \int_0^d I_{im}(r) r dr$, where $r = (x^2 + y^2)^{1/2}$ and d is the radius of the integration (experimentally limited by the size of the CCD matrix). To calculate energy inside the diaphragm in the Z-scan configuration, we must integrate over a circular aperture with radius $r_a = \omega_i [\ln(1/(1-S))]^{1/2}$, where S is the closed aperture linear transmission and ω_i is the beam waist in the image plane in linear regime. The latter is physically equal to the beam radius at the entry ($\omega_i = \omega_e$) because the magnification of considered 4f imaging system is unity. Thus, the closed aperture Z-scan energy is evaluated through the integral $\varepsilon_D = 2\pi \int_0^{r_a} I_{im}(r) r dr$, while the energy outside the opaque disc in the EZ-scan configuration must be calculated with $\varepsilon_D = 2\pi \int_{r_a}^d I_{im}(r) r dr$. Using the latter quantities, we define diffraction efficiency separately for nonlinear and linear regimes:

$$\eta = \frac{\varepsilon_{D_{NL}} - \varepsilon_{D_L}}{\varepsilon_{T_{NL}} - \varepsilon_{T_L}}, \quad (3)$$

where $\varepsilon_{D_{NL}}$ and ε_{D_L} are the energies inside the aperture or outside the opaque disc in nonlinear and linear regimes, respectively, whereas $\varepsilon_{T_{NL}}$ and ε_{T_L} denote the total energies in the image plane in nonlinear and linear regimes, respectively. The subtraction of low irradiance background Z-scan ($\varepsilon_{D_L}/\varepsilon_{T_L}$) from the high irradiance scan ($\varepsilon_{D_{NL}}/\varepsilon_{T_{NL}}$)

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