Invited Paper

Dark-field Z-scan imaging technique

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We report on Dark-Field Z-scan (DFZ-scan) as a new imaging technique combining Z-scan method with Dark-field microscopy in order to measure optical refraction nonlinearity. Numerical and experimental results are provided to validate this concept. The image of the induced phase shift is spatially resolved without introducing a complex interferometric setup. Moreover, the experimental results show almost 3 times increase of the sensitivity when compared to the conventional Z-scan method. New perspective of microscope laser scanning is introduced.

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1. Introduction

There are many nonlinear (NL) optical characterization techniques with proper advantages and inconveniences. Starting with the degenerated four-wave mixing [1] which only gives the modulus of the nonlinear susceptibilities, reaching gradually nonlinear imaging technique [2] combining simplicity and elegance with Z-scan method [3,4], Z-scan is able to distinguish the real from the imaginary part of the nonlinear optical susceptibility. It is found to be the most widely used technique in this field nowadays. Since its apparition so far it is being improved in a continuous way through Eclipsed Z-scan [5], Dual-arm Z-scan [6], and D4e-Z-scan [7]. Other efficient techniques have emerged, briefly we will only mention: the beam deflection measurement technique [8] and the Scattered Light Imaging Method (SLIM) [9] showing that new methods to characterize NL response of the material are still relevant and subject to active research, in order to find simple and sensitive ways to measure, especially, the induced nonlinear refraction at high intensity despite the presence of relatively high NL absorption. Other original characteristics include two stacked samples to be measured in a single run [10] where one material could be used as a reference.

The purpose of this paper is to demonstrate the feasibility of a new optical measurement technique based on dark field microscope configuration combined with Z-scan method. The experimental acquisitions are performed and compared with related numerical simulations. The use of an annular illumination light and a magnified image of the focal plane, where the NL material is moving, provides new characteristics in the measured signal. The dark field microscopy leads to a dark background image on which the observation of small structures stands out, clearly increasing the contrast in the NL diffracted energy. One can thus obtain images of the induced phase shift spatially resolved without requiring the introduction of a complex interferometric technique [11]. The gain in sensitivity and intrinsic advantageous using this method are specified.

2. Principle of the measurement and theory

The Dark-field microscopy and Z-scan method are mature and popular techniques. The result of their combination will be labeled as DFZ-scan. The setup used to implement the DFZ-scan is presented in Fig. 1.

The system is composed of two subsystems: the first one with lens L1 (20 cm focusing length) is used to focus an annular illumination delivered by the object \(O(x,y)\) into the sample positioned at \(z\) in the focal region, the second subsystem is used to obtain a magnified image onto the CCD. This imaging subsystem is composed of lens L2 (10 cm focusing length) positioned in such a way as to obtain a 5x magnification by adjusting the distance \(d\) between the focal plane of L1 and lens L2 to be equal to 12 cm; while the distance \(D\) between L2 and the CCD, has been fixed to 60 cm (see Fig. 1). Fixed to L2 a circular aperture C is inserted where the radius is adjusted in such a way as to block the direct rays coming from the annular illumination (see Fig. 1). The sample (NLM) is mounted on a translation stage and moved along the beam direction (z axis) around the focal point. For each step of the motor (1 mm), one image is recorded by a CCD camera. A second arm is used to monitor the energy fluctuation of the incident laser...
propagation is performed on a distance \( z' = f \) from the object plane to \( L1 \). Then we propagate the beam up to the sample located at \( z \) using \( z' = f + z \) in the optical transfer function \( H \). Next, the NL response of the material is taken into account, before continuing the propagation with the same formalism up to the image plane of \( L2 \) considering \( z' = d - z \) before this lens, and \( z' = D \) after it.

We assume cubic nonlinearity and a thin NL medium of thickness \( L \) exhibiting (i) linear absorption defined by \( a(m^{-1}) \), (ii) two-photon absorption defined by \( \beta(m/W) \) and (iii) NL refraction defined by \( n2(m^2/W) \). In these conditions, the transmission of the sample is described by

\[
T(u, v, z) = \left\{ \left[ 1 + q(u, v, z) \exp(\alpha L) \right]^{1/2} \exp(j\Delta\phi_{\text{eff}}(u, v, z)) \right\}.
\]

where \( q(u, v, z) = \beta L_{\text{eff}} I(u, v, z) \) with \( L_{\text{eff}} = [1 - \exp(-\alpha L)]/\alpha \), and \( I(u, v, z) \), related to the input electric field \( E(u, v, z) \), denotes the intensity of the laser beam within the sample. The NL phase shift is \( \Delta\phi_{\text{eff}}(u, v, z) = 2\pi n2 L_{\text{eff}} I(u, v, z)/\lambda \), in which we define the effective intensity as \( I_{\text{eff}}(u, v, z) = I(u, v, z)\ln[1 + q(u, v, z)]/q(u, v, z) \). To characterize more simply the nonlinearities we use the on-axis at the focus NL absorption \( q_0 = \beta L_{\text{eff}} I_0 \) and the NL phase shift \( \Delta\phi_0 = \Delta\phi_{\text{eff}}(0, 0, 0) \), where \( I_0 \) is the focal on-axis intensity.

3. Experimental details and numerical simulation

Before starting the DFZ-scan experiments we used the 4f system [15] which was aligned carefully to obtain a magnification equal to 1, in such a way that it was possible to characterize accurately the profile of the annular beam at the entry in the linear regime (Fig. 2). The image of the annular home-made object is registered in the computer allowing to take into account the real profile of the object when necessary, and to measure the size of the object to adjust the radius of \( C \). The dimensions of the annular object gave the following results: the mean value of \( R_{e} \), the inner radius of the disc, was equal to 0.73 mm and \( R_{e} \), the outer radius, was 1.58 mm (Fig. 2). These values are used in the simulation defining the object as

\[
O(x, y) = \text{circ} \left( \frac{\sqrt{x^2 + y^2}}{R_e} \right) - \text{circ} \left( \frac{\sqrt{x^2 + y^2}}{R_i} \right).
\]

Fig. 1. Dark field Z-scan imaging system. The sample (NLM) is scanned along the beam direction around the focal plane \((z = 0)\). The labels refer to: annular illumination \((O)\), circular aperture \((C)\), lenses \((L1, L2 \text{ and } L3)\), beam splitters \((BS1 \text{ and } BS2)\), CCD camera \((CCD)\) and mirrors \((M1 \text{ and } M2)\). (For interpretation of the references to color in this figure text, the reader is referred to the web version of this article.)

Fig. 2. Image of the real annular object used to illuminate the NL material \([t1(x,y)]\). This image is obtained using a 4f system [7]. \( R_e \) and \( R_i \) are the mean values of the annular object radiuses.

pulses (via lens \( L3 \)). While moving the sample, the self-diffraction on the induced NL phase-shift (and/or absorption) pattern originates changes in the transmitted beam. A part of this beam (blue dashed line in Fig. 1) passes through the circular aperture \( C \) centered on lens \( L2 \), which act as a spatial filter blocking the direct light (red solid line in Fig. 1) and preventing direct optical rays from reaching the CCD, thus obtaining relatively dark image in the absence of any nonlinearity. In our experiment, the material is moved along \( z \) (the optical axis) to vary the intensity inside the material. When the incident intensity is high enough, self-diffraction of the light on the induced phase spatial modulation in the nonlinear material is imaged through the lens \( L2 \). Then, measuring the change of the diffracted light in the image plane allows the detection of a characteristic signal related to nonlinearities.

Theoretically, the whole system is described with basis on Fourier optics [12]. The general scheme of beam propagation inside the system is similar to that described in [7], [13] and [14], where we must pay attention to the lens \( L2 \) which is not positioned in the same way as in the usual 4f system. To summarize the beam propagation and for self-consistency, let us consider \( E(x,y) \) as the amplitude field at the entry of the setup, \( x \) and \( y \) being the spatial coordinates. At the output of the annular aperture, the field is described as \( O(x,y) = E(x,y) * t1(x,y) \), where \( t1(x,y) \) is the transmittance of this aperture. The spatial spectrum of the object is given by

\[
\hat{O}(u,v) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} O(x,y) \exp[-j2\pi(ux + vy)] dx dy.
\]

where \( \hat{O} \) denotes the Fourier transform operation, \( u \) and \( v \) are the normalized spatial frequencies. In general, the field is propagated over a distance \( z' \) by taking into account the transfer function of the wave propagation phenomenon

\[
H(u,v,z') = \exp \left\{ j2\pi \left[ 1 - (\lambda u)^2 - (\lambda v)^2 \right] \right\},
\]

where \( \lambda \) is the wavelength. Then the field amplitude at \( z' \) is obtained by computing the inverse Fourier transform \( E(x,y,z') = \hat{O}^{-1}[S(u,v)H(u,v,z')] \).

To calculate the output beam after passing through a lens with focal length \( f \), we apply the phase transformation related to the thickness variation: \( t1(x,y) = \exp[-j\pi(x^2 + y^2)/f] \). The first
the function $\text{circ}(r/R)$ is defined equal to 1 if the radius $r$ is less than or equal to $R$ and 0 elsewhere.

Experiments were done with a block of 3.6 mm thick fused silica plate. Silica is a material known as relatively difficult to measure because of its low $n_2$ value ($\sim 10^{-20}$ m$^2$/W). Acquisitions were performed using an Nd: YAG laser emitting linearly polarized light at 532 nm (12 ps, FWHM pulse duration) at a repetition rate of 10 Hz. To reduce the thermally generated noise attributable to dark current, we used a CCD camera (14 bits) with a cooling system stabilizing the temperature sensor at 0°C, having 1380 x 1104 area of 9.08 µm x 9.08 µm square pixels. The circular aperture $C$ situated just before the imaging lens $L_2$ (Fig. 1) had a radius equal to 0.25 mm (imposed by the drilling) which was chosen smaller than the theoretical maximal value given by simple rays tracing: $r_c = dR/\phi_c \approx 0.44$ mm.

Fig. 3 shows the comparison between the simulated result (b) using the procedure detailed in section II and the experimental image (a) obtained under the same conditions (at $z=0$). The qualitative agreement is very good, the images are almost identical, presenting the same size.

For each experiment, two sets of acquisitions are performed. The first set is in the NL regime and the second by reducing the incident laser intensity in order to operate in the linear regime. This is necessary to remove from the NL acquisitions the diffraction, diffusion and/or imperfection due to the sample inhomogeneities. We perform a Z-scan from $-30$ mm to $+30$ mm with a 1 mm step. So for each scan we register 61 images on the hard disk. After taking into account the reference beam to correct the energy fluctuations, we integrate over all pixels of the diffracted image (as the one shown in Fig. 3) to calculate the total transmitted energy.

Fig. 4 shows the signal of the diffracted energy (filled circle points) versus $z$, the position of the NLM together with the obtained numerical simulation (blue solid line). The normalization is done with respect to the diffraction in the linear regime, giving a signal equal to 1 when the NLM is far from the focus. It was considered the following experimental parameters: $L=3.60$ mm, $n_2=3 \times 10^{-20}$ m$^2$/W, $I_0=55$ GW/cm$^2$ (giving $\Delta \phi_0=0.68$ rad), $I_0$ is measured after calibration considering the value of $n_2=3 \times 10^{-18}$ m$^2$/W for CS$_2$ given in reference [4]. The numerical simulation obtained with these parameters is shown together with the experimental data. Note the very good agreement between the simulation and the experimental acquisition. A maximum is followed by a minimum for positive self-focusing indicating the ability to determine the positive sign of the nonlinearity. Of course a negative $n_2$ will result in an opposite Z-scan trace characterized by a minimum followed by a maximum. In order to obtain simple relationship relating the NL phase shift with the signal, simulation has been performed (see Fig. 5), using the same parameters and only varying the induced NL phase shift to calculate the diffracted energy in the image plane versus $z$.

Also the energy diffracted in the NL regime is normalized to that obtained in the linear regime. Obviously, the signal increases with the incident intensity but more specifically, it is the difference $\Delta T_{pv}$ between the maximum and the minimum which could be used for the measurement as for conventional Z-scan. The study of $\Delta T_{pv}$ based on the nonlinear phase shift at the focus $\Delta \phi_0$ shows a quadratic dependence (see Fig. 6). A quadratic fit of this dependence (blue dashed curve) gave us the following relation: $\Delta T_{pv} = \Delta \phi_0 \times (0.64 \times \Delta \phi_0 + 0.61)$, which is valid between 0 and 1.5 rad.

This relationship allows us to accurately determine $\Delta \phi_0$ by measuring $\Delta T_{pv}$ in order to obtain the NL coefficients. But a more simple linear relationship is preferable by performing a linear fit (solid red line) which allows to estimate this phase shift inside the useful interval where usually the measurements are done: for $0.2 < \Delta \phi_0 < 1.3$ rad we obtain $\Delta T_{pv} = 1.3 \times \Delta \phi_0 - 0.12$. This relationship can be used to estimate the NL phase shift within 20% accuracy. According to the definition of the sensitivity as the ratio
this new method is estimated to be 3.2 times more sensitive than that of the original Z-scan method where the equivalent relation is \( \Delta \phi = 0.41 \times \Delta \phi_0 \) for the on-axis closed aperture trace. We performed also experimental comparison of this signal with that obtained by the conventional Z-scan, using the same sample under the same conditions (12 ps, 532 nm, and rectilinear polarization). Fig. 7 shows the usual closed aperture Z-scan signal obtained at 26 GW/cm\(^2\) with a linear transmission of the closed-aperture Z-scan equal to 33%.

Comparing the signal obtained in Fig. 4 at 55 GW/cm\(^2\) \((\Delta T_{pv} = 0.7)\) and the signal obtained here (Fig. 7, \(\Delta T_{pv} = 0.14\)) at almost half of this intensity, it is found that the experimental gain in sensitivity is approximately equal to \((26 \times 0.7)/(55 \times 0.14) \approx 2.3\). This value is close to the theoretical value of 3.2 obtained with the on-axis Z-scan transmittance (linear transmittance of the closed aperture \(\sim 0\)) because more precisely we have to correct this obtained gain in sensitivity taking into account the linear transmittance of 33% considered in the experiment related to Fig. 7. The gain that would be obtained with a completely closed ideal aperture should be \(2.35/(1 - 0.33)^{0.25} = 2.6\) when using the result of the numerical fitting obtained in [4] relating the signal \(\Delta T_{pv}\) to the NL phase shift. This validates our theoretical calculation on the sensitivity demonstrating a gain of a factor almost equal to 3. Of course, higher sensitivity could be obtained with other methods as in [5,8], but the benefits and opportunities of this technique are numerous. The numerical simulations show that it is possible to obtain a signal whose response is mainly due to NL refraction whatever the NL absorption is (see Fig. 8). There is no need to divide the closed aperture Z-scan with the open one. Indeed, performing numerical simulation with the same parameters as those involved in Fig. 4, but adding a relatively high NL absorption \((q_0 = 1.9)\), we obtained approximately the same signal. Further application of this property is on the way to characterize high NL absorbing materials as those found in porphyrin compositions to improve the reliability of the NL refraction measurements [16]. Moreover, the image given on the CCD when the sample is in the focal plane is closely related to the spatial phase shift extension induced in the sample, which allows us to measure the size of the...
output beam with an appropriate magnification, and to prevent the collapse of the beam when the operator notices a high variation of the size. Previously, this spatial resolution was obtained using interferometric techniques, as in [11] where the setup was difficult and complex to align and the processing was tedious to perform. When higher orders NL measurements are to be carried out [7,13], it will be more convenient to visualize directly the beam diameter at the output of the NL material than to deduce from the diffracted far field in the CCD plane a possible collapse of the beam, as it has been done up to now using the 4f system or Z-scan setups.

When compared to eclipsed Z-scan (EZ-scan) [5] which is known to be very sensitive, an advantage of this new method is that it does not require a perfectly Gaussian incident beam. Moreover, the enhancement of the sensitivity in EZ-scan comes at the expense of a reduction in accuracy. Here also the size of the spatial filter C and its influence on the sensitivity is not provided because the use of a reference material for calibration is highly recommended and the purpose of this paper remains principally restricted to demonstrate the feasibility of the method. In order to illustrate the behavior of a highly nonlinear material such as CS$_2$ which is frequently used as a reference material to calibrate the setup we performed an experiment using 1 mm thick cell at $I_0=9$ GW/cm$^2$ under the same experimental conditions. CS$_2$ is known to have a high nonlinear response when compared to silica.

Fig. 9 shows this behavior, the data in blue filled squares represent the signal obtained with CS$_2$ (1 mm thick cell at $I_0=9$ GW/cm$^2$). Red points are the acquisitions shown in Fig. 4 (3.6 mm thick silica plate at 55 GW/cm$^2$). The blue dashdot line and the red solid line are the simulation taking into account the experimental parameters.

Fig. 9. Comparison of the diffracted energy in the image plane versus $z$ for high and low nonlinear material. Data in blue filled squares represent the signal obtained with CS$_2$ (1 mm thick cell at $I_0=9$ GW/cm$^2$). Red points are the acquisitions shown in Fig. 4 (3.6 mm thick silica plate at 55 GW/cm$^2$). The blue dashdot line and the red solid line are the simulation taking into account the experimental parameters.

4. Conclusion

In summary, the dark-field Z-scan method is based on the measurements of the non-direct diffracted light that passes through a circular aperture placed in the far-field region just before a lens magnifying the image of the output NL material. Z-scan is achieved using dark-field images with a relatively higher contrast. The experimental acquisition and the numerical simulation show very good agreement and demonstrate gain in sensitivity 3 times higher than conventional Z-scan. This imaging technique is also advantageous in presence of NL absorbing material where the signal is almost related to the induced NL refraction only. It allows to prevent collapse for high order NL coefficient estimation and to develop further potential applications in laser scanning microscopy.

References


