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D4 σ nonlinear measurement inside a 4f-Z-scan system

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Abstract: The D4 σ method using a 4f-Z-scan system is presented in order to increase the sensitivity and the resolution in Z-scan measurements. The numerical calculations and the experimental results validate our approach.

The inspection of linear and nonlinear (NL) optical parameters such as refractive index and absorption coefficient is essential for most of the applications in optics (optical limiting, lasers, optical amplifiers, etc.). Measurements of NL refraction and absorption using the Z-scan method [1] are widely performed nowadays. A variant of this method called Baryscan [2] reporting an increased phase distortion resolution (PDR) ($\lambda/50\,000$) has been published using stable CW laser input. More recently van Stryland's group reported a dual-arm Z-scan technique [3] improving the PDR up to $\lambda/1\,000$ using pulsed lasers. These recent studies show that improvement of the PDR is still relevant. Measuring the diffraction efficiency allowed us to compare the sensitivity of different techniques inside a Z-scan 4f-system [4]. But still remain open questions to fully understand the physical phenomenon contributing for sensitivity and PDR improvements.

Fig. 1. (a) Schematic of the 4f-system. The sample (M) is moved along the focal region. The labels refer to: lenses (L_1 - L_2). (b) Relative variation of the beam waist versus z . The other parameters are $\phi_{NL0}^{eff} = 0.8$ and $q_0 = 0.6$; (c) Calculated $\Delta\omega_{pv}$, versus the effective phase shift.

We demonstrate here that the sensitivity of Baryscan is twice lower than that of Z-scan and we show that the use of hard-(physical) apertures (as a razor blade) with pulsed laser presenting pointing fluctuations could be a severe limitation leading to both lower sensitivity and small S/N ratio when compared to the direct measurement of the output beam waist variation using a CCD sensor.

The NL image formation inside the 4f -system shown in fig. 1 (a) is described using a model based on Fourier optics. The general scheme of beam propagation inside the 4f-system is described in details in [4]. We assume that the electric field at the object plane is Gaussian. We propagate the beam using the Helmholtz wave equation and the phase transformation related to lens thickness variation up to the image plane, taking into account the NL response of the material in the focus. We assume cubic nonlinearity and a thin NL medium of thickness L exhibiting (i) linear absorption defined by α (m^{-1}), (ii) two-photon absorption defined by β (m/W) and (iii) NL refraction defined by n_2 (m^2/W). We define the on axis NL parameters at $z = 0$, $q_0 = \beta L_{eff} I_0$ and $\phi_{NL0}^{eff} = 2\pi n_2 L_{eff} I_0^{eff} / \lambda$ as the NL absorption and the effective phase shift, respectively. Here $L_{eff} = (1 - e^{-\alpha L}) / \alpha$, I_0 denotes the on-axis intensity within the sample and $I_0^{eff} = I_0 \log[1 + q_0] / q_0$, the effective intensity. The beam waist measurement is performed using the ISO standard definition [5]. Based on the second moment of $I(x, y)$ the D4 σ method gives 4 times the standard deviation of the intensity distribution. The beam radius in the x direction is: $\omega_x = 2\sqrt{\int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} I(x, y) (x - \bar{x})^2 dx dy / \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} I(x, y) dx dy}$ where $\bar{x} = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} I(x, y) x dx dy / \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} I(x, y) dx dy$ is the centroid of the beam. For Gaussian beams, the D4 σ

method gives the same result as the $1/e^2$ method. The calculated $(\omega_{NL}-\omega_L)/\omega_L$, the relative variation of the beam waist versus z , the position of the sample in the focus, is shown in Fig. 1 (b), where ω_{NL} and ω_L denote respectively the mean values (measured along x and y) of the beam waist associated to the NL and linear profiles at the output of the 4f-system. The difference between the peak and the valley $\Delta\omega_{pv}$ shown in Fig. 1 (c), is a function of the effective phase shift at the focus for a Gaussian input beam showing a linear relation ($\Delta\omega_{pv} = 0.34 \times \phi_{NLO}^{eff}$). Moreover the linearity remains valid in presence of relatively high NL absorption.

Fig. 2. (a) Comparison between D4σ (squares) with that of Baryscan (circles) profiles. The solid and dashed lines show the calculations. (b) Comparison of the beam waist relative variation (squares; vertically shifted to 1) and the usual normalized Z-scan transmittance (stars) for a highly NL absorbing material. The inset shows the open aperture Z-scan transmittance. The NL parameters obtained were: $q_0 = 0.6$ (see the inset in Fig. 2b) and according to Fig. 1, $\phi_{NLO}^{eff} = 0.2$.

The detailed description of our experimental setup is given in [4]. Excitation is provided by a Nd:YAG laser linearly polarized 17 ps single pulses at $\lambda = 1064 \mu m$. In the image plane we use a CCD camera performing Z-scan, Baryscan and D4σ profiles out from the same acquired images at each z -position by numerically changing the soft aperture. When using the D4σ method one have to calculate the centroid of the beam inside the frames which is equivalent to follow the pointing instability of the pulsed laser and thus reduces the noise that could appear with hard apertures as the ones used with Z-scan and Baryscan.

Fig. 2 (a) shows the experimental results for As_2Se_3 , a chalcogenide glass with relatively high NL absorption. Notice that the Baryscan signal is totally buried inside the noise due to laser pointing instability while the beam waist measurement method gives a higher signal to noise ratio. The same behaviour could be found for Z-scan using hard aperture. The calculated profiles (solid line for D4σ and dashed line for Baryscan) are shown in the same figure. The agreement is very good. Moreover the D4σ method is unambiguously twice more sensitive than the Baryscan method because physically, we consider the totality of the pixels in order to measure the beam waist. So the quantity of information is twice that obtained with the razor blade in Baryscan. This is illustrated in Fig. 2 (a) by the difference between the peaks and the valleys inside the two scans processed for the same acquisitions showing approximately a factor 2 in the sensitivity. Moreover the PDR in Baryscan using pulsed laser with hard aperture in our experiment is approximately $\lambda/180$ which is a factor 5 lower than the PDR obtained with the D4σ method.

To obtain the comparison between Z-scan and D4σ and to improve the signal-to-noise ratio in Z-scan the centroid calculation is used for both profiles in Fig. 2(b) to correct for pointing laser fluctuations. The processing have been made using the same acquired images showing unambiguously the same sensitivity. The radius of the soft circular closed aperture in Z-scan is calculated to maximize the optical diffraction efficiency [4]. However, the advantage in the D4σ method when compared to Z-scan is that there is no need to divide two different Z-scan traces in order to obtain the NL refractive response.

In summary, the D4σ method is insensitive to pointing instability of the pulsed laser because no hard aperture is employed as in the Z-scan or Baryscan methods. Numerical calculations allow to obtain simple relations that can be used for the measurements simplifying the procedure especially for NL absorbing material.

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