# Single shot interferometric method for measuring the nonlinear refractive index

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**Abstract:** In this paper we are introducing a new single shot method for measuring the nonlinear refractive index of materials in a simple interferometric pump-probe configuration. The theoretical model proposed for extracting the nonlinear refractive index from the experimental fringe pattern and the experimental configuration are presented and discussed. The results obtained by this method are in good agreement with that obtained on the same sample using the conventional Z-scan method.



**OCIS codes:** (190.0190) Nonlinear optics; (120.3180) Interferometry.

## References and links

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## 1. Introduction

The modification of the refractive index of a material due to intense light illumination is one of the most studied nonlinear optical effects [1]. The measurement of the nonlinear refractive index, an important material parameter that characterizes the nonlinear refractive response,

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has significant implications in photonics applications as optical switching, routing, limiting, processing, solitons [1, 2]. Over the years many methods have been proposed for this type of characterization [2]. Basically, in most methods a light beam (excitation beam) induces a refractive index change in the investigated material, which, in turn, changes the phase of a probe beam. By measuring the induced phase change, the nonlinear refractive index can be determined. For example, the wave mixing methods [3] involve the use of at least two mutually coherent laser beams to write a refractive index grating. The diffraction efficiency of the induced grating is monitored with a probe beam, which can be different from the writing beams or it can be one of the writing beams (self-diffraction). In wave mixing, time resolved pump-probe techniques are used to determine the time constants of the processes responsible for the nonlinear response of the material. Using these methods is not possible to determine the sign of the nonlinear refractive index change. The complexity of the experimental setup is also an inconvenient of these methods. Other method for measuring the nonlinear refractive index is the Z-Scan method [4, 5] with its derivatives (I-Scan [6], reflection Z-Scan [7–9], etc.). Being a single beam technique, relatively simple to implement, the Z-Scan method is a widely used techniques in our days for nonlinear optical measurements, providing the magnitude and the sign of the optical nonlinearity. An inconvenient of conventional Z-scan consists in the lack of information on the time constants of the involved nonlinear processes. These time constants can be determined using pump-probe Z-scan, but this is more difficult to implement.

In this paper we are proposing a new, simple, pump-probe technique for the measurement of the light induced refractive index change. This method consists on using an interferometric technique to measure the phase change induced in a probe beam when passing through a nonlinear medium excited with a pump beam. In this method, a single interference image is sufficient to extract the phase change from which the nonlinear refractive index can be determined. We describe the theory on which is based this method and the experimental configuration. We experimentally tested this method and the results obtained are in good agreement with that obtained using the Z-Scan method on the same sample [10]. We also discuss the advantages of this method.

## 2. Description of the method

We consider a Gaussian excitation beam incident on a nonlinear sample at an angle with the projections on two orthogonal axes (X, Y) given by $\theta_{e,x}$ and $\theta_{e,y}$, respectively:

$$I(x,y,z=0)=I_0\cdot e^{\left(-\frac{\left((x-x_0)\cdot\cos(\theta_{e,x})\right)^2+\left((y-y_0)\cdot\cos(\theta_{e,y})\right)^2}{w^2}\right)}. \tag{1}$$

In this expression, $w$ is the radius of the beam, centered at $x_0$ and $y_0$, at the incidence on the sample ($z = 0$). In the simplest case, when only third-order nonlinearities are excited, the change of the refractive index in the sample is given by:

$$\Delta n(x,y,z)=n_2\cdot I_{ex}(x,y,z), \tag{2}$$

in which $n_2$ is the nonlinear refractive index due to the third-order nonlinearities and

$$I_{ex}(x,y,z)=I(x,y,z=0)\cdot e^{-\alpha_0\cdot z}. \tag{3}$$

It can be seen that the refractive index change follows the intensity profile of the incident beam. We consider beam diffraction negligible during the propagation through the sample thickness. This condition is satisfied when the sample thickness is much smaller than the Rayleigh length of the beam (thin samples). For a sample thickness one order of magnitude smaller than the Rayleigh length, the nonlinear phase change difference due to diffraction is less than 0.4%. For thick samples or strong focusing of the excitation beam, diffraction should be considered. Due to the sample absorption, characterized by the absorption

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coefficient $\alpha_0$, a decreasing refractive index change is induced along the thickness of the sample.

The nonlinear phase change induced by this refractive index change to a probe beam is given by:

$$\Delta\Phi_{nl}\left(x,y,z=L\right)=k\cdot\int_0^L\Delta n\left(x,y,z\right)\cdot dz. \tag{4}$$

Here, $k$ is the wave number of the probe beam and $L$ is the thickness of the sample. By substituting Eqs. (2) and (3) into Eq. (4) and solving the integral we obtain at the output of the sample:

$$\Delta\Phi_{nl}\left(x,y,z=L\right)=k\cdot n_2\cdot I_{ex}\left(x,y,z=0\right)\cdot\frac{1-e^{-\alpha_0 L}}{\alpha_0} \tag{5}$$

In this equation one can observe that the excitation beam will induce a phase modulation of the probe beam that will follow the transversal intensity profile of the excitation beam. We consider that the nonlinear phase change is acquired only during the propagation of the probe beam through sample and suffers no other change until the observation plane:

$$\Delta\Phi_{nl}(x,y)\equiv\Delta\Phi_{nl}\left(x,y,z>L\right)=\Delta\Phi_{nl}\left(x,y,z=L\right). \tag{6}$$

We can use the same procedure in the case when nonlinearities of higher orders are also excited in the sample. For example, in the case when third- and fifth-order nonlinearities are involved, the light induced nonlinear phase change can be written as a sum of terms corresponding to nonlinearities of different orders and is given by [11]:

$$\Delta\Phi_{nl}\left(x,y,z=L\right)=\Delta\Phi\left(x,y,z\right)^{(3)}+\Delta\Phi\left(x,y,z\right)^{(5)}+\cdots \tag{7}$$

with:

$$\Delta\Phi\left(x,y,z=L\right)^{(3)}=kn_2 I_{ex}\left(x,y,z=0\right)\left[1-\exp\left(-\alpha_0 L\right)\right]/\alpha_0, \tag{8-a}$$

$$\Delta\Phi\left(x,y,z=L\right)^{(5)}=kn_4 I_{ex}^2\left(x,y,z=0\right)\left[1-\exp\left(-2\alpha_0 L\right)\right]/2\alpha_0. \tag{8-b}$$

In these equations $n_2$, $n_4$, are the nonlinear refractive indices corresponding to third- and fifth-order nonlinearities, respectively.

In the case of saturated nonlinear optical refraction, the nonlinear refractive index change is given by:

$$\Delta n\left(x,y,z\right)=\frac{n_2\cdot I_{ex}\left(x,y,z\right)}{1+I_{ex}\left(x,y,z\right)/I_{sat}}. \tag{9}$$

Here $I_{sat}$ is the saturation intensity of the nonlinear refractive index change. If we use this in Eq. (4) we obtain:

$$\Delta\Phi_{nl}\left(x,y,z=L\right)=\frac{k\cdot n_2\cdot I_{sat}}{\alpha_0}\cdot\ln\left[\frac{I_{sat}+I_{ex}(x,y,z=0)}{I_{sat}+I_{ex}\left(x,y,z=0\right)\cdot e^{-\alpha_0\cdot L}}\right] \tag{10}$$

Knowing the excitation intensity $I_{ex}$ and the sample properties, $\alpha_0$ and $L$, one can obtain the nonlinear refractive indexes from the measured nonlinear phase change by using one of the previous Eqs. (5), (7), or (10).

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*Phase change extraction from a single interference image*

To measure the induced phase change we are using a Michelson interferometer due to its simple design and availability. This Michelson interferometer is using a laser at a wavelength that will not excite the sample nonlinearities. The sample is placed in one of the interferometer arms at normal incidence. The interferometer is slightly misaligned in order to produce linear fringes, which are easier to analyze. The intensity pattern given by the Michelson interferometer is:

$$I(x,y)=I_1(x,y)+I_2(x,y)+2\sqrt{\left(I_1(x,y)\cdot I_2(x,y)\right)}\cdot\cos\left(\Delta\Phi(x,y)\right). \tag{11}$$

Considering the double pass of the probe beam through the sample, the phase difference between the two interfering beams is given by:

$$\Delta\Phi(x,y)\equiv\Delta\Phi_{total}(x,y)=2\cdot\Delta\Phi_{nl}(x,y)+\Delta\Phi_{tilt}(x,y), \tag{12}$$

where

$$\Delta\Phi_{tilt}(x,y)=k\cdot\left[x\cdot\sin\left(\theta_x\right)+y\cdot\sin\left(\theta_y\right)\right], \tag{13}$$

is the phase difference introduced by the tilt of the beams in the interferometer placed in air. Here, $\theta_x$ and $\theta_y$ are the projections of the tilt angle on two orthogonal planes, transversal to the propagation direction.

To experimentally obtain the value of the nonlinear refractive index $n_2$ we have to extract the phase difference from interference images. We use the Fourier Transform Method (FTM) [12, 13] to extract the phase difference, $\Delta\Phi_{total}(x,y)$, from a single interference image. Other methods for direct spatial reconstruction of optical phase (DSROP) [13] can be also used. Following [12–14] the FTM is briefly described below. It consists of filtering the phase modulated image in the Fourier domain in order to obtain the phase modulation given by the nonlinear response of the sample. Substituting Eq. (12) and Eq. (13) into Eq. (11) and considering parallel fringes with $\theta_y \approx 0$ we obtain

$$I(x,y)=A(x,y)+B(x,y)\cdot\cos\left(2\cdot\Delta\Phi_{nl}(x,y)+k\sin(\theta_x)x\right), \tag{14}$$

where

$$A(x,y)=I_1(x,y)+I_2(x,y)\ \text{ and }\ B(x,y)=2\sqrt{I_1(x,y)+I_2(x,y)}.$$

We can rewrite Eq. (14) as:

$$I(x,y)=A(x,y)+C(x,y)\cdot e^{ikx\sin(\theta_x)}+C^*(x,y)\cdot e^{-ikx\sin(\theta_x)}, \tag{15}$$

where

$$C(x,y)=(1/2)\cdot B(x,y)\cdot e^{2i\Delta\Phi_{nl}(x,y)} \tag{16}$$

and * denotes the complex conjugate. Taking the Fourier transform (denoted with ~symbol) of Eq. (15) we obtain:

$$\tilde{I}(u_x,u_y)=\tilde{A}(u_x,u_y)+\tilde{C}(u_x-k\sin\left(\theta_x\right),u_y)+\tilde{C}^*(u_x+k\sin\left(\theta_x\right),u_y), \tag{17}$$

where $u_x$ and $u_y$ are spatial frequencies (coordinates in the Fourier space). To extract the nonlinear phase modulation, the second term in Eq. (17) (or, equivalently, the third) must be used. This term can be easily extracted in the spatial frequencies domain if the separation of the terms is good, without overlapping of different Fourier terms. This can be done by increasing the main spatial frequency, i.e. increasing the beam tilt in the *x* direction. By

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translating the filtered component in the center of the spectrum and taking the inverse Fourier transform, we obtain Eq. (16). From this equation it is easy to obtain the nonlinear phase change distribution, $\Phi_{nl}(x,y)$, wrapped in a $2\pi$ interval. To obtain a continuous phase distribution, without $2\pi$ phase jumps, one can apply different phase unwrapping algorithms [15, 16]. By fitting the resulted phase distribution with one of the Eqs. (5), (7), or (10) we can obtain the value of the nonlinear refractive index.

## 3. Experimental demonstration of the method

As previously mentioned, our experimental setup is based on a Michelson interferometer with the sample introduced in one of its arms, Fig. 1(a). The pump laser is a frequency-doubled Nd:YAG laser operating in continuous wave (c.w.) regime at 532 nm wavelength. The laser is focused to a 0.48 mm spot having the on-axis intensity of 84 W/cm$^2$ at the sample plane. For an easy comparison with existing Z-Scan data, in these experiments we have used the same laser and one of the samples as in our previous work [10]. A cell with colloidal CdTe nano-crystals in water, with the size chosen to have the first excitonic resonance near the excitation laser wavelength is the test sample used in the experimental demonstration of the method.

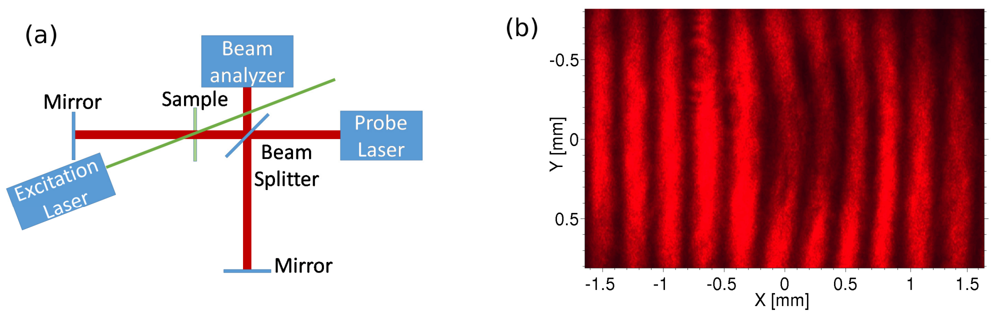


Fig. 1. The experimental setup based on a Michelson interferometer (a) and an experimental fringe pattern obtained when the investigated sample is excited (b).

The probe laser is a c.w. laser at 633 nm. To be absolutely sure that the probe laser will not induce optical nonlinearities in the sample we have performed Z-Scan experiments on the sample using it as excitation laser and no refractive index change was observed. In Fig. 1 one can observe the slight tilt of the excitation laser beam, as previously mentioned in the theoretical section (see Eq. (1)). A normal incidence of the excitation laser beam on the sample would have imposed the use of a beam splitter in the respective arm of the interferometer with further distortions of the probe beam and complicating the data analysis. Using FTM, previously described, to analyze the acquired interference pattern shown in Fig. 1(b), we derived the induced phase difference shown in Fig. 2.

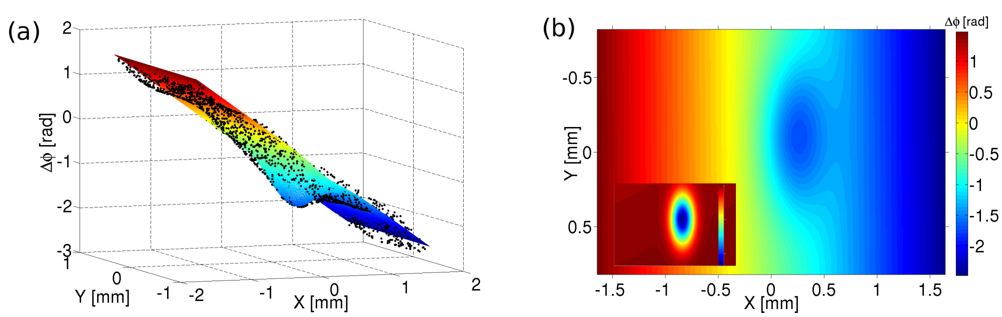


Fig. 2. (a) 3D representation of the reconstructed phase (dots) fitted with Eq. (10) (surface). (b) The top view of the fitting surface. Inset: the reconstructed phase change corresponding to the tilted Gaussian excitation only.

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In Fig. 2, one can observe the general slope of the phase due to the tilt of the two beams from the interferometer arms. The tilted incidence of the excitation beam on the sample, with respect to the normal one, deforms the transversal shape of the Gaussian phase change, corresponding to the excited area of the sample, from circular to an elliptic one (inset in the Fig. 2(b)). Both influences are considered in the final fitting function, as described in the theoretical section, and can be clearly observed in Fig. 2. Fitting the experimental data with the theoretical function that uses the saturated nonlinearity (Eq. (10) we obtained the value for the nonlinear refractive index, $n_2 = -16\cdot10^{-7}$ cm$^2$/W and for the saturation intensity, $I_{sat} = 700$ W/cm$^2$ (Fig. 2, right), in agreement with the values obtained by Z-Scan in a previous work, using the same sample (sample E in [10], $n_2 = -14.65\cdot10^{-7}$ cm$^2$/W and $I_{sat} = 674.5$ W/cm$^2$).

## 4. Discussions and comments

In comparison with wave mixing methods, this method is using a single pump beam for excitation, reducing the complexity of the experimental setup and, in addition, provides the sign of the nonlinear refractive index change. As in the wave mixing methods the temporal dynamics of the nonlinear processes is easy to investigate by this pump-probe method. Compared to the Z-Scan technique the experimental setup used in this method has no moving parts, the same area of the sample is excited during the entire experiment and experiments with single shot excitation can be implemented. In this method the correlation of the scanning parameters with the dynamics of the nonlinear processes as in Z-Scan and I-Scan methods is avoided that leads to an easier interpretation of the experimental data.

A single interference pattern obtained with a single shot exposure contains all the information needed for the characterization of the nonlinear response excited in the sample (different orders of nonlinearity, saturation of nonlinearity, etc). This makes the proposed method robust for the characterization of nonlinear optical materials.

The sensitivity and accuracy of the method is limited mainly by the algorithm for phase extraction from an interference image. Due to the great improvement of today image sensors, the FTM was used in many applications with sub-nanometer accuracy [17]. The method sensitivity is given by $2\pi/N$, where N is the number of pixels (px) in the direction orthogonal to the fringe direction [16]. To have a quantitative estimate, for N = 1280 px the minimum detectable phase change is of the order of 0.005 rad ($\sim\lambda/1000$). Considering the excitation intensity of the order of 1 GW/cm$^2$ at $\lambda = 1\mu$m in a sample of 1 mm thickness, the minimum measurable nonlinear refractive index is of the order of $10^{-16}$ cm$^2$/W.

When high temporal resolution is needed, a single pass (e.g. Mach-Zehnder) interferometer should be used. The use of a double pass interferometer doubles the sensitivity but also decreases the signal to noise ratio that could reduce the accuracy. However, noise reduction algorithms provide very good results in improving the signal to noise ratio [18].

## 5. Conclusion

We have proposed a simple and easy to implement method for the measurement of the nonlinear refractive index. In this pump-probe method, from a single interference pattern it is possible to extract all the information needed for the characterization of the nonlinear refractive response excited in the sample. The method was experimentally tested and the results are in good agreement with the Z-Scan results on the same sample.

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