

## Review

# Measurement of nonlinear refraction of thick samples using nonlinear-imaging technique with a phase object



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## ABSTRACT

The third-order nonlinear refractive index of carbon disulfide (CS<sub>2</sub>) with different thicknesses is measured by a nonlinear-imaging technique with a phase object (NIT-PO). The propagating characteristics of the laser beam within thick medium are analyzed by numerically solving the electromagnetic wave equation. By theoretically calculating, we study the relationship of the phase shift induced by nonlinear refraction versus medium thicknesses. Fitting the output image with the help of the effective interaction length, the nonlinear refractive index  $n_2$  of CS<sub>2</sub> can be determined, which suggests a one-single-shot method for the investigation of optical nonlinearity in thick medium.

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## 1. Introduction

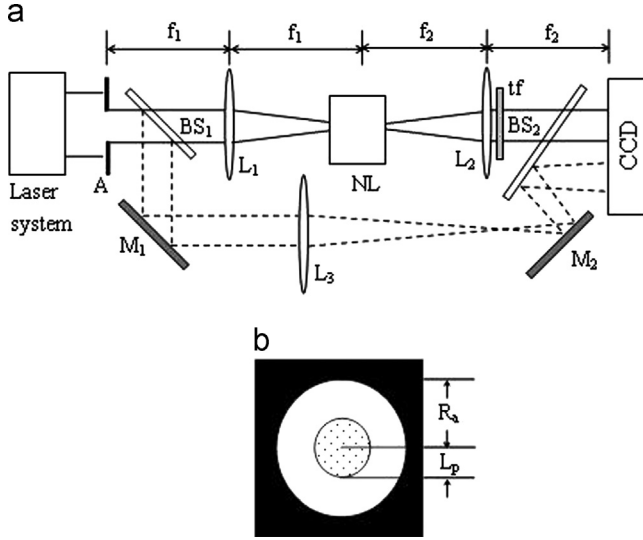
In 1996, Boudebs et al. introduced a nonlinear-imaging one-laser-shot technique based on the 4f coherent imager system to measure the nonlinear refraction of nonlinear optical materials placed at the Fourier plane of the 4f setup. This technique, however, can not distinguish the sign of third-order nonlinear index [1]. Subsequently, an improvement was adopted: a circular diaphragm was placed at the entrance of 4f system [2], and

simultaneously added a quarter-wave-length phase object (PO) in the object plane of the 4f imaging system, which, like the Z-scan method [3], can expediently and simultaneously obtain the sign and amplitude of the nonlinear refractive index [4]. This improvement can increase the sensitivity of the measurement significantly. The 4f coherent imaging technique with phase object has several advantages comparing to other measurement methods [5,6] such as no movement of sample, simple optical setup and one-single-shot measurement. Nevertheless, the previous studies are nearly focused on thin sample (the thickness of the medium is much smaller than the Rayleigh diffraction length of laser beam). As known to all, for a thin sample, the relationship between the nonlinear phase shift and effective length of the material can be expressed as:  $\Phi_{NL} = kn_2L_{eff}I$  ( $k$  is the wave vector,  $n_2$  is

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**Fig. 1.** (a) Schematic of 4f coherent system imager. The center of nonlinear material (NL) is located in the focal plane. The aperture and the CCD camera are at the entry and exit plane of the 4f system respectively. A, aperture with a phase object; L<sub>1</sub>–L<sub>3</sub>, lenses; M<sub>1</sub>, M<sub>2</sub>, reflecting mirrors; BS<sub>1</sub>, BS<sub>2</sub>, beam splitters; tf, neutral filter. (b) Schematic of the circular aperture with phase object at the entry of the 4f system. The inner circular region is uniform phase-shift area by depositing SiO<sub>2</sub> on the glass plate at a certain thickness.

third-order nonlinear refractive coefficient,  $L_{eff}$  is the effective length of the medium, and  $I$  is the intensity in the nonlinear material, respectively). The way to get greater nonlinear phase shift is either increasing the light intensity or the thickness of the sample. Without doubt, the above relation will be questionable when the thin sample approximation is dissatisfied. Some studies have been conducted on the optical nonlinear properties of thick sample using Z-scan technique [7–16] and a simple transmittance measurement technique with a phase object (T-PO) [17], and the theoretical model and some analytical results were obtained. So far, to our knowledge, the investigation on the optical nonlinearity of thick medium using NIT-PO has not been reported.

In this paper, the third-order nonlinear refractive index of CS<sub>2</sub> is investigated with different thicknesses by using NIT-PO. By numerically simulating, the relationship of phase shift induced by nonlinear refraction versus medium length can be obtained. The average value of nonlinear refraction index with the different sample thickness is determined.

## 2. Theoretical model and discussion

A typical arrangement of the 4f coherent imaging system is shown in Fig. 1(a). The circular aperture with a phase object is illuminated at normal incidence by a linearly polarized monochromatic plane wave (defined by  $E = E_0(t)\exp[-j(\omega t - kz)] + c.c.$ , where  $\omega$  is the angular frequency,  $k$  is the wave vector, and  $E_0(t)$  is the amplitude of the electric field containing the temporal envelope of the laser pulse) emitted by a pulsed laser system. By considering a top-hat beam, the circular aperture at the entrance of the 4f system defines the object with transmittance:  $t_a(r) = \text{circ}(r/R_a)$  (where  $r$  is the radial coordinate at the entrance of 4f system and  $R_a$  is the radius of aperture). At the center of the aperture, there is a circular phase object with radius  $L_p$  ( $L_p < R_a$ ) which has a uniform phase retardation  $\phi_L$ . So the transmittance of the total aperture (with PO) can be written as:  $t(r) = \text{circ}(r/R_a) + (\exp(j\phi_L) - 1)\text{circ}(r/L_p)$ . Consequently, the field at the rear plane of the aperture with a phase object is  $E_1(r, t) = E(r, t)t(r)$ . The field in the front surface of lens L<sub>1</sub> for

aperture with a phase object placed at a distance equals to the focal length  $f_1$  of L<sub>1</sub> can be obtained by using the Fresnel diffraction integral

$$E_2(r_1, t) = \frac{2\pi}{i\lambda f_1} \exp\left(\frac{i\pi r_1^2}{\lambda f_1}\right) \int_0^{+\infty} r dr E_1(r, t) \exp\left(\frac{i\pi r^2}{\lambda f_1}\right) J_0\left(\frac{2\pi r r_1}{\lambda f_1}\right), \quad (1)$$

where  $r_1$  is the radial coordinate in this plane,  $J_0$  is the Bessel function of the first kind of zero-order. The phase transformation due to the lens L<sub>1</sub> in the paraxial approximation is expressed by  $t_{L_1} = \exp(-ikr_1^2/2f_1)$ . The amplitude of the field in the rear plane of L<sub>1</sub> can be written as:  $E_2' = E_2 t_{L_1}$ . The second propagation is performed on a distance  $d_1 = f_1 - L/2$  ( $L$  is the thickness of the sample), the field amplitude in the front surface of nonlinear material is defined as

$$E_3(r_2, t) = \frac{2\pi}{i\lambda d_1} \exp\left(\frac{i\pi r_2^2}{\lambda d_1}\right) \int_0^{+\infty} r_1 dr_1 E_2'(r_1, t) \exp\left(\frac{i\pi r_1^2}{\lambda d_1}\right) J_0\left(\frac{2\pi r_2 r_1}{\lambda d_1}\right), \quad (2)$$

with  $r_2$  is the radial coordinate in the plane of L<sub>1</sub>. Using the slowly varying envelope approximation (SVEA) and beam propagation method (BPM) [19,20], the field within the nonlinear medium can be written as [21–23] (the detail process is presented in the Appendix)

$$2jk_0 n_0 \frac{\partial E_4(r_2, z, t)}{\partial z} = \nabla_{\perp}^2 E_4(r_2, z, t) - jk_0 n_0 \alpha E_4(r_2, z, t) + k_0^2 (n^2(r_2, z, t) - n_0^2) E_4(r_2, z, t), \quad (3)$$

where  $E_4(r_2, z, t)$  is the electric field envelope of the light in the sample,  $\nabla_{\perp}^2$  is the transverse Laplace operator,  $z$  is the propagation distance in the sample,  $k_0 = \omega/c$  is the wave vector,  $\alpha = \alpha_L + \alpha_{NL}$  is the total optical absorption coefficient, which consisted of the linear absorption coefficient  $\alpha_L$  and the nonlinear absorption coefficient  $\alpha_{NL}$ ,  $n = n_0 + \Delta n$  is the refractive index distribution in material,  $n_0$  and  $\Delta n$  is the linear refractive index and nonlinear refractive index, respectively. By solving Eq. (3), the electric field  $E_4(r_2, L, t)$  of the rear surface of the nonlinear material can be obtained.

The next propagation is performed on a distance  $d_2 = f_2 - L/2$ , and the amplitude of the field in the front of lens L<sub>2</sub> can be obtained by using the Fresnel diffraction

$$E_5(r_3, t) = \frac{2\pi}{i\lambda d_2} \exp\left(\frac{i\pi r_3^2}{\lambda d_2}\right) \int_0^{+\infty} r_2 dr_2 E_4(r_2, L, t) \exp\left(\frac{i\pi r_2^2}{\lambda d_2}\right) J_0\left(\frac{2\pi r_3 r_2}{\lambda d_2}\right) \quad (4)$$

with  $f_2$  is the focal length of L<sub>2</sub> and  $r_3$  is the radial coordinate in this plane, respectively. The phase change due to the lens L<sub>2</sub> in the paraxial approximation is expressed by  $t_{L_2} = \exp(-ikr_3^2/2f_2)$ . The amplitude of the field in the rear plane of L<sub>2</sub> can be written as:  $E_5' = E_5 t_{L_2}$ . The final propagation is conducted at a distance  $f_2$ , which is the image plane, and the field amplitude is expressed as

$$E_0(r_0, t) = \frac{2\pi}{i\lambda f_2} \exp\left(\frac{i\pi r_0^2}{\lambda f_2}\right) \int_0^{+\infty} r_3 dr_3 E_5'(r_3, t) \exp\left(\frac{i\pi r_3^2}{\lambda f_2}\right) J_0\left(\frac{2\pi r_0 r_3}{\lambda f_2}\right). \quad (5)$$

the fluence recorded by the CCD camera which placed at the output plane of the 4f system can be written as

$$F = \int_{-\infty}^{+\infty} |E_0(r_0, t)|^2 dt \quad (6)$$

Using Eqs. (1)–(6), one can numerically calculate the image at the output of the 4f system, which is detected by the CCD camera. In Fig. 1(a), the branch split by the beam splitter BS<sub>1</sub> and then collected by L<sub>3</sub> to the CCD camera used to monitor the energy fluctuation of the input laser pulse.

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