Simple measurement of nonlinear refraction by using a single pulse and beam*

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A very simple technique, by which both the magnitude and the sign of nonlinear refraction can be determined through using only one single pulse and beam with a phase object, is presented. Using this technique, only the transmittance of an aperture in the far field is investigated. We study the nonlinear refraction of the carbon disulfide by using the presented technique with 21 ps pulses at a wavelength of 532 nm as a test.

Keywords: nonlinear refraction, phase object

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1. Introduction

Various techniques have been reported for the measurement of the nonlinear refraction in materials, such as nonlinear interferometry,[1,2] degenerate four-wave mixing,[3] Z-scan,[4] and the nonlinear-imaging technique with a phase object (NIT–PO).[5] Thereinto, Z-scan is widely used for determining nonlinear parameters of various materials because of its some salient features such as simplicity and high sensitivity.[6] However, in this technique the sample needs moving and multiple measurements are required during the materials passing through the focal plane under a strong laser pulse excitation. This can lead, in some cases, to the damage of samples. The single-shot technique would overcome the above difficulties. NIT–PO is based on the 4f coherent imaging system to characterize the nonlinear optical properties of materials. Compared with the conventional Z-scan technique, NIT–PO technique has several advantages, such as simple alignment, no displacement of the nonlinear medium, one single laser shot for estimating the nonlinear refraction, lower sensitivity to statistical fluctuations of the laser beam. But several images must be managed and this process is relatively complicated. On the side, the high demand for the CCD induces the expensive cost of instruments in this technique. Recently, we have reported on a PO Z-scan technique to distinguish the transient thermal lensing effect from the third-order nonlinear refraction.[7,8] The PO Z-scan trace of the pure negative (positive) third-order nonlinearity shows a single valley (peak) shape. In the present paper, we propose a very simple technique, which is a simplified PO Z-scan technique, to investigate the nonlinear refraction of the sample.

2. Experiment setup and theoretical analysis

The single pulse and beam transmittance with a phase object (SPBT–PO) configuration is shown in Fig. 1, which is very similar to the one of the PO Z-scan technique. The only difference between these two setups is that no displacement of the sample is required in SPBT–PO technique, but it is needed in the PO Z-scan technique. The sample is fixed near the focal plane of the lens. A PO, as shown in Fig. 1(b), locates at the front focal plane of the lens. In the far field, an aperture with the radius (r_a) equal to the radius of diffraction transmission beam inside the PO is arranged. The branch split by the beam splitter BS, detected by detector D_1, is used to record the energy of input laser pulse. The transmitted energy of the aperture is measured by detector D_2. The transmittance (D_2/D_1) is measured under two different con-

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ditions, one is linear condition (low intensity), and other is nonlinear condition (high intensity). Dividing the transmittance measured under the nonlinear condition by the one measured under the linear condition, we can obtain the normalized nonlinear transmittance, which is related to the nonlinear refraction of the sample.

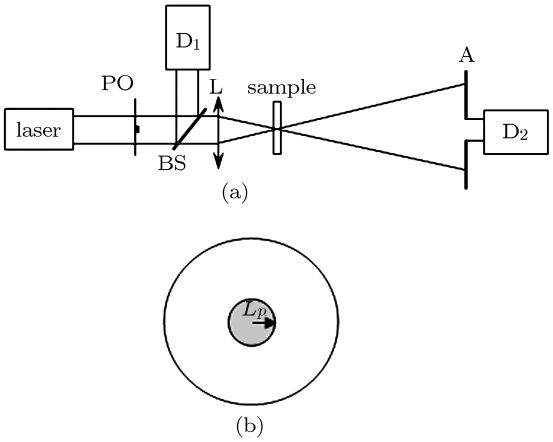


Fig. 1. (a) Schematic of SPBT–PO system. PO: phase object, L: lenses, BS: beam splitters, D_1 and D_2: energy detector. (b) Schematic of the phase object. The inner circular area is the uniform phase shift area which is of SiO_2 deposited on glass plate with a certain thickness.

The waist of linearly polarized Gaussian beam, ω_e, at the entry of the setup can be expressed as

$$E(r,t) = E_0 \exp\left[-\frac{r^2}{\omega_e^2}\right] \exp\left[-\frac{t}{2\tau^2}^2\right], \qquad (1)$$

where r is the transverse polar coordinate; E_0 denotes the amplitude of the electric field of the laser pulse; τ is the laser pulse width (FWHM).

The circular PO of radius L_p having a uniform phase shift φ_L is placed at the centre of the Gaussian beam. The transmittance of the object can be written as $t(r) = \exp(\mathrm{i}\,\varphi_L)$ if $r < L_p$ and 1 elsewhere. The amplitude of the field at the output of the PO is $E_{01}(r,t) = E(r,t)t(r)$. The amplitude of the field in the front surface of the sample placed at the focal plane of the lens L can be obtained by the Fourier–Bessel transform

$$E_{02}(r_1,t) = \frac{2\pi}{\lambda f}\int_0^\infty rE_{01}(r,t)\mathrm{J}_0(2\pi r r_1)\mathrm{d}r, \qquad (2)$$

where f is the focal length of lens L, r_1 is the radial coordinate in this plane. J_0 is the Bessel function of the first kind of zero order.

For simplicity, only the third-order nonlinear refraction is investigated here. Considering a thin medium and using the slowly varying envelope approximation yield the equations of the phase and the irradiance for a beam propagating in the sample, respectively, as follows:

$$\frac{\mathrm{d}\Delta\phi}{\mathrm{d}z'} = k\gamma I, \qquad (3)$$

$$\frac{\mathrm{d}I}{\mathrm{d}z'} = -\alpha I, \qquad (4)$$

where $\Delta\phi$ represents the phase change, and z' is the propagation length in sample; γ is the nonlinear refraction coefficients; α is the linear absorption coefficient; I is the intensity in the sample, $I = |E_{02}|^2$ at $z' = 0$.

The amplitude of the field rear surface of the sample can be written as

$$E_{03}(r_1,t) = E_{02}(r_1,t)\exp(-\alpha L)\exp\left[\mathrm{i}\,\Delta\phi(r_1,t)\right]. \qquad (5)$$

The amplitude of the field at aperture A can be achieved with the Fresnel diffraction integrals, and it is given as

$$E_a(r_2,t) = \frac{2\pi}{\mathrm{i}\,\lambda d}\exp\left(\frac{\mathrm{i}\,\pi r_2^2}{\lambda D}\right)\int_0^{+\infty} r_1\mathrm{d}r_1 E_{03}(r_1,t) \times \exp\left(\frac{\mathrm{i}\,\pi r_1^2}{\lambda D}\right)\mathrm{J}_0\left(\frac{2\pi r_2 r_1}{\lambda D}\right). \qquad (6)$$

Similarly, in the linear case, the field of the aperture can also be obtained, and defined as $E'_a(r_2,t)$. Here, D is the distance between the aperture and the focal plane; r_2 is the radial coordinate in this plane. While the integration of irradiance over the pulse duration and the aperture cross section gives an energy corresponding to that detected by D_2. Then the normalized nonlinear transmittance T can be calculated to be

$$T = \frac{\int_{-\infty}^{+\infty}\int_0^{r_a} 2\pi r_2 |E_a|^2 \mathrm{d}r_2\mathrm{d}t}{\int_{-\infty}^{+\infty}\int_0^{r_a} 2\pi r_2 |E'_a|^2 \mathrm{d}r_2\mathrm{d}t}. \qquad (7)$$

The nonlinear refraction coefficient γ can be deduced by fitting the numerical calculated nonlinear transmittance to the experimental nonlinear transmittance. Meanwhile, if T is larger than 1, it implies that the sign of the nonlinear refraction is positive, otherwise, it is negative.

3. Results and discussion

Carbon disulfide (CS_2) is chosen as the sample in our experiment because it is a standard sample. A Q-switched and mode-locked Nd:YAG 532 nm laser

(EKSPLA, PL2143B) with a pulse width of 21 ps (FWHM) and repetition rate of 2 Hz, is used. Its spatial and temporal distributions are both nearly Gaussian. The sample solution is placed in a 2-mm-thick quartz cell. The focal length of the lens is $f = 400$ mm. The PO consists of a glass plate on which a transparent dielectric disc of radius $L_p = 0.5$ mm is deposited. The disc has a thickness and index of refraction such that it retards the phase of the incident light by $\phi_L = \pi/2$ at 532 nm. The distance between the aperture and the focal plane is $D = 1.6$ m, and the radius of the aperture is $r_{\rm a} = 2$ mm. The waist of the Gaussian beam at the entry of SPBT–PO system is $\omega_{\rm e} = 2.9$ mm, giving rise to a beam waist at the focal plane $\omega_0 = 23$ μm. The energy of the input laser pulse is 0.22 μJ.

The sample is placed first in the far field to obtain the linear transmittance, and then it is moved to the focal plane of L to obtain the nonlinear transmittance. Under our experimental conditions, the normalized nonlinear transmittance T is measured to be 1.4481, which is larger than 1, and it indicates that the sign of the nonlinear refraction of CS_2 is positive. By varying the value of γ to obtain the best fitting between the numerically calculated nonlinear transmittance and the experimental nonlinear transmittance, the value of γ for CS_2 is obtained to be $n_2 = 3.3 \times 10^{-18}$ m^2/W, which is in good agreement with results presented by previous reports.

4. Conclusion

We report on a powerful but very simple technique to characterize the nonlinear refraction coefficient. By using this technique, both the sign and the amplitude of the nonlinear refraction coefficient can be measured. The optical configuration is very easy to realize and needs no displacement of the material. It requires only one-laser-shot. Therefore, the cost of the measurement is considerably reduced accordingly.

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