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New holographic technique for third-order optical properties measurement

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Abstract

We present a new nonlinear holographic method, which yields the real and imaginary parts of the third-order susceptibility ($\chi^{\langle 3\rangle}$) of an optical medium. The method consists in producing a nonlinear interference pattern between two beams of different intensity, where the strong beam induces nonlinear effects in the tested sample. Only three laser shots are required to extract both real and imaginary parts of $\chi^{\langle 3\rangle}$ by means of the images recorded on a CCD camera. We apply the method to measure the nonlinear refractive index (n_2) of new synthesized organic materials in the pico-second regime (30 ps pulse duration at 1064 nm). The obtained results show that the proposed technique allows the measurement of n_2 coefficients in diluted materials with a sensitivity of $\lambda/1500$ at 1 GW cm^{-2} of input intensity.

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1. Introduction

Materials with large optical nonlinearities have been extensively investigated due to their many potential applications in integrated optics [1], all-optical switching [2] and optical processing information [3]. They are generally characterized by their nonlinear absorption coefficient (β) and the nonlinear refractive index (n_2), which are respectively used to evaluate the imaginary (Im $\chi^{\langle 3\rangle}$) and real (Re$^{\langle 3\rangle}$) parts of the third-order susceptibility $\chi^{\langle 3\rangle}$. Numerous techniques have been proposed for measuring with high sensitivity n_2 and

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β coefficients [4–9]. Among them, Z-scan technique is one of the most versatile ones [5]. This method consists in detecting changes of the wave fronts of a single beam in the far field when the nonlinear medium is scanned through the focal plane of the beam. The sign and magnitude of n_2 can be deduced by plotting the transmittance of a small aperture as a function of the sample position. Despite its sensitivity and experimental simplicity, Z-scan has limitations in measuring n_2 of materials with low nonlinear optical response, because it requires high incident intensity and averaging over many laser pulses for each sample position, experimental conditions, which can lead to sample damage. As an alternative to Z-scan, the I-scan method has been recently proposed [10,11], which consists in using two closed Z-scan configurations. Fixing the position of two samples (n_2-reference and n_2-test) at the respective focal plane of each Z-scan arm and taking simultaneously the measurements of the transmission in the far field, the nonlinear refractive index can be obtained from the slope of the experimental curve $T_r = f(T_t)$, where T_r and T_t are, respectively, the normalized transmittances of the reference and test samples. Using this method, n_2 values of the order of 10^{-20} cm^2/W could be measured in non-homogenous materials and diluted solution media [10]. However, this method has also the limitations similar to the usual Z-scan ones.

Recently, a new Mach–Zehnder (MZ) interference technique for measuring third-order susceptibility has been proposed [12]. Basically, the method consists of a pump–probe experiment in which the wave front of a weak probe beam is changed by nonlinearity induced by the strong incident pump beam. The first arm of the MZ interferometer is used as the reference beam and the second arm as the probe beam where the sample is located. The interference patterns are recorded on a CCD camera and the strong intensity in the pump beam induces changes in the interference pattern. In this way, the nonlinear variation index can be deduced using only one laser shot.

In this work, we develop a new experimental holographic method for measuring n_2 in materials with poor nonlinear optical properties. The method is a variant of the MZ-interference technique where image-processing procedures are used in order to estimate n_2 from the recorded holographic patterns. The measurements are validated by using the standard nonlinear liquids: carbon disulfide (CS_2) and benzene (C_6H_6). The method is also tested by measuring low n_2 coefficients of some materials such as chloroform ($CHCl_3$) [13] and methanol solution of organic salts of carboxilate anions with chiral amines and ammonium cations [14]. The obtained results are compared with the ones reported in a previous work [10], where the same organic materials were measured by using I-scan.

The advantages of the proposed method are simple alignment and high sensitivity (up to λ/1500), which allows measuring n_2 and β coefficients quickly without damaging the samples when high incident intensities are used due to the fact that only three laser shots are required for extracting the final results.

2. Theoretical consideration

We consider the schematic diagram shown in Fig. 1, where two beams ⟨1⟩ and ⟨2⟩ impinge onto two identical nonlinear media. We also suppose that the intensity of the beam ⟨1⟩ is sufficiently strong for inducing third-order nonlinear effects in the sample S_1 and that the intensity of the beam ⟨2⟩ is weaker than the beam ⟨1⟩, such that none nonlinear effects are produced in the sample S_2.

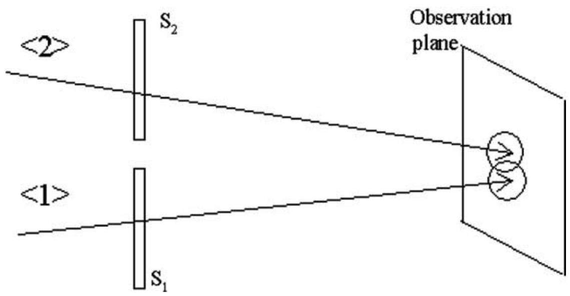

Fig. 1. Schematic diagram explaining the parameters used in the theory.