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Experimental evaluation of inhomogeneous nonlinear refractive index distribution using time-resolved inline digital holography

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Abstract: Time-resolved inline digital holography was validated during study of nonlinear optical properties of graphene microparticles on the sample glass. A set of probe pulse inline digital holograms were recorded and compared with numerically simulated data. © 2021 The Author(s)

1. Introduction

By now, several methods were proposed for the study of nonlinear optical phenomena. The development of such approaches is motivated by the rapid growth of interest in the development and manufacturing of novel photonic and optoelectronic devices based on nonlinear optical phenomena [1]. One of such phenomena is the photorefractive effect [2], described by the dependence of the dielectric polarization of the material on the light intensity. When considering this type of optical nonlinearity, the coefficient of nonlinear refractive index n2 is an important parameter for characterizing nonlinear media or photonic devices, and the development of novel techniques aimed at its evaluation is becoming the subject of close attention of scientists. Although evaluation of this optical characteristic can be performed using for example z-scanning technique, the method is not adapted to the measurement of local inhomogeneities of the nonlinear refractive index of the sample. Meanwhile study of such samples is of considerable interest due to the rapid development and utilizing of layered structures [2] and composite materials [3] in science and technology. Recently time-resolved inline digital holography (TRIDH) approach for investigation of the optical samples with constant nonlinear refractive index was proposed [4]. The method was also validated on layered samples and the samples with embedded inhomogeneities of nonlinear refractive index in numerical experiments reported in [5, 6]. In this work, we present the preliminary results of experimental monitoring of a transparent glass sample with deposited graphene particles, which nonlinear refractive index is significantly different from one of the substrate.

2. Experimental setup and inline digital holograms recording

Time-resolved inline digital holography approach is based on detection and analysis of the probe pulse inline holograms after its noncollinear degenerate phase modulation (NDPM) inside the sample during its propagation along with femtosecond pump pulse. According to the experimental setup (see Fig. 1(a)) the two femtosecond laser pulses propagate inside the sample at a small angle, while the delay line of the pump pulse allows to vary the positions of the two pulses overlay area inside the sample. During propagation inside the sample, a small phase shift is induced to the probe beam within the overlay area with pump pulse due to significant pump pulse power density and nonlinear refractive index n_2 of the studied sample as demonstrated in figure 1(b). In the case of inhomogeneous distribution of nonlinear refractive index inside the sample (presence of local inhomogeneities of nonlinear refractive index (LINRI)) variation of the nonlinear optical parameter results in a change of the phase shift induced to the probe beam. Propagation of the probe pulse from the object to the image plane results in the formation of the diffraction pattern, which can be further analyzed by comparison with numerically simulated intensity distributions according to the developed numerical NDPM model [5,6]. In this study, a sample glass with graphene particles deposited onto its surface was studied using the described approach. By variation of the pump pulse delay line horizontal scanning of the sample was performed and local diffraction patterns generated due to NDPM of the probe beam on graphene microparticles were observed in different areas of the sample. Figure 1(c) demonstrates three fragments of typical diffraction patterns observed at different pump pulse delays, which contain slightly distorted concentric diffraction rings. Numerical processing of the obtained data in accordance with TRIDH approach suggests simulation of similar diffraction patterns generated by NDPM on various simulated nonlinear inhomogeneities and their fitting to experimentally obtained patterns.

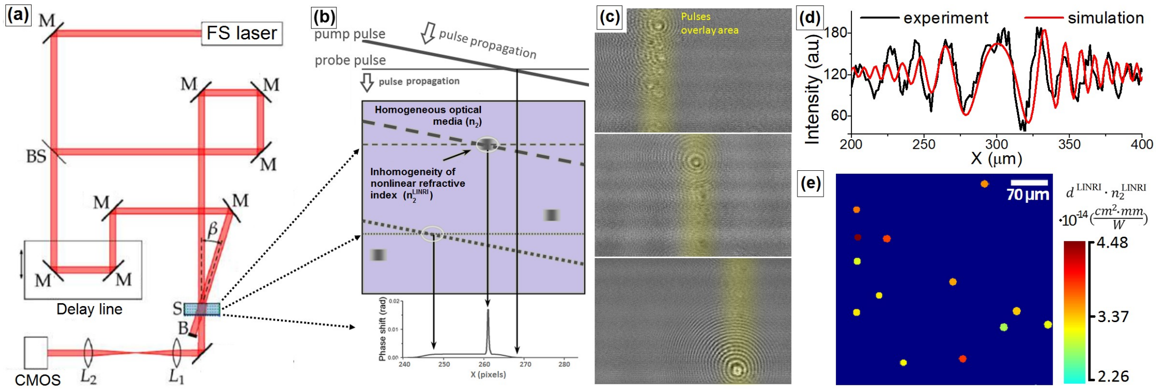

Fig. 1. Schemes of: (a) experimental setup utilized in TRIDH study of inhomogeneous optical object and (b) generation of probe pulse phase shift profile. Examples of: (c) experimentally recorded probe pulse diffraction patterns and (d) experimental pattern cross-section fitted by numerically simulated data. Figure (e) shows a fragment of evaluated nonlinear optical properties of microparticles deposited onto glass substrate.

3. Data processing and evaluation of nonlinear optical properties of inhomogeneous sample

In contrast with the simplest case of constant nonlinear refractive index, in the case of inhomogeneous n_2 distribution comparison of the experimentally recorded diffraction patterns with numerically simulated data is much more difficult due to a large number of parameters to be varied. These parameters include nonlinear refractive index of the substrate n_2, nonlinear refractive index of local inhomogeneities of nonlinear refractive index n_2^{LINRI}, their thickness d^{LINRI} and position [x^{LINRI}, y^{LINRI}] as well and temporal parameter τ^{LINRI}, characterizing relaxation of the induced variation of dipole moment by pump pulse. In addition, the aberrations of the optical system, unknown location and depth of local inhomogeneties of nonlinear refractive index (LINRI), incorrect estimation of the distance between the object and image planes and pump pulse power density lead to inaccurate numerical simulation of the experimentally obtained results. In the case of homogeneous media with a constant refractive index [4], an almost perfect approximation of the experimentally obtained diffraction pattern using only two parameters (n_2 and τ) is possible. In the case of an inhomogeneous distribution of the nonlinear refractive index, a complete iterative fit of the experimental data requires a routine change of many parameters, which makes the exact solution of the problem very difficult. However approximate fitting of the experimentally recorded intensity distributions with numerically simulated data is still possible, although its quality is lower than that demonstrated in our previous study [4].

An example of the numerical fitting of experimental diffraction patterns cross-sections is demonstrated in figure 1(d). Similar fitting was performed during the analysis of several dozens of LINRI and nonlinear optical properties of the studied sample were evaluated. Due to the fact that the thickness of the LINRI d^{LINRI} was unknown, the proposed allowed us for evaluation of the product of thickness and nonlinear refractive index $d^{LINRI} \cdot n_2^{LINRI}$ rather than nonlinear refractive index of the inhomogeneities themselves. An example of the reconstructed location and color-coded $d^{LINRI} \cdot n_2^{LINRI}$ of several LINRIs in a small area of the studied sample is presented on figure 1(e). Despite imperfect fitting of the experimental data with numerically simulated data we believe that the proposed method can be useful for the experimental study of modern optical materials and samples with inhomogeneous nonlinear refractive index.

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4. References

[1] Kauranen M., Zayats A.V. "Nonlinear plasmonics." *Nature photonics* **6**, 737-748 (2012).
[2] Autere A., et al. "Nonlinear optics with 2D layered materials." *Advanced Materials* **30**, 1705963 (2018).
[3] Belashov A.V., et al. "Relative variations of nonlinear elastic moduli in polystyrene-based nanocomposites." *Polymer Testing* **95**, 107132 (2021).
[4] Petrov, N. V., et al. "Time-resolved inline digital holography for the study of noncollinear degenerate phase modulation." *Optics Letters* **43,** 3481-3484 (2018).
[5] Belashov A.V., et al. "On the Possibility of Studying the Fraction of Inhomogeneities in Optical Nonlinear Media Using Time-Resolved Inline Digital Holography." *Technical Physics* **66,** 822–830 (2021).
[6] Belashov A.V., Cheng C.J., Petrov N.V. "Noncollinear degenerate phase modulation in samples with inhomogeneous optical nonlinear properties [Invited]." *Applied Optics* **60,** B14-B22 (2021).