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Application of Photothermal Digital Interferometry for Nonlinear Refractive Index Measurements within a Kerr Approximation

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ABSTRACT

The methodology of Photothermal Interferometry implemented through off-axis digital holography for the nonlinear refractive index measurements of optical media with the thermal mechanism of nonlinearity is presented. An experimental appraisal is done on the example of chlorophyllin 1% solution in ethyl alcohol. It allows us to estimate the effective value of nonlinear refractive index as $-0.65 \cdot 10^{-3}$ cm^2/W. The comparison of the experimental result with data obtained by means of a reference approach was performed. Possible errors lead to a mismatch between them are highlighted and analyzed.

1. Introduction

Self-action effects occur in the process of light propagation with high enough intensity through an optically nonlinear medium. They consist in the changes of original medium properties under the influence of this incoming radiation. The optical nonlinear media or any nonlinear optics phenomena are increasingly used in the various spheres of human activity. The development of spectroscopic techniques [1], implementation of new materials for power limiting [2], control of thermal blooming and laser filamentation effects in atmosphere [3,4], invention of the new methods of optical superresolution [5] or the new approaches to increase the field of view of optical systems [6], hidden image recovery [7] or numerical light propagation in optical nonlinear media [5,8,9] are only the few examples where the knowledge of nonlinear optics principles is on demand.

At the present time many different types of optical nonlinear media are known and this list is constantly growing. In each of these media some nonlinear mechanisms prevail under certain conditions that determines their practical characteristics important in various applications. There are several methods to measure optical nonlinear properties, the most popular among them to our knowledge is Z-scan technique [10,11] (and its modifications, for example, I-scan or P-scan [12,13], reflection Z-scan [14,15] and etc.) or the approaches based on degenerate four-wave mixing (DFWM) [16]. Currently, there is the wide range of nonlinear optical spectroscopy methods [17] which can be used to study optical nonlinear properties. They provide the measurement of a nonlinear absorption coefficient, the nonlinear change of refractive index or other nonlinear optical properties of investigated media. These include such techniques as polarization or stimulated emission spectroscopy and many other approaches. The variety of techniques based on degenerate or nondegenerate four wave mixing (NDFWM) [17] (p. 431, 445) can also be attributed to the field of nonlinear optical spectroscopy. In addition, interferometric measurements are often applied to the estimation of nonlinear refractive index [18,19].

The mechanism of optical nonlinearity can be caused by various physical effects, which determines not only the nonlinear characteristics of the medium but also the requirements for the methods used to measure these nonlinear properties. For example, there are electronic-vibrational, thermal, photorefractive and other mechanisms of optical nonlinearity. It is well known that optical media with electronic-vibrational mechanism has a response time by orders of magnitude less in comparison with the media of thermal

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type or the photorefractive mechanism of nonlinearity. On the other hand their nonlinear refractive index is also much smaller, than corresponding value for the optical nonlinear media of thermal type. Many papers reports were concerned with the application of time-resolved interferometry techniques for the study of optical media with the primarily electronic-vibrational mechanism of nonlinearity [18–22]. If the optical nonlinearity mechanism of some medium has a thermal origin, a high temporal resolution is not required. The application of an interferometric approach remains relevant, since refractive index variance can be measured by the detection of phase distributions changes caused by a thermal lens effect. However in order to consider the thermal lens effect to be similar with a Kerr effect one should fulfill several requirements, associated mainly with the presence of convection processes.

In this paper we describe the application of Photothermal digital interferometry (PI) for the measurements of the nonlinear refractive index of optical media with the thermal mechanism of nonlinearity. Much attention is paid to the discussion of accepted assumptions, the analysis of obtained experimental results and its comparison with the indirect approach based on an analytical equation derived in [23]. The validation of the described technique is performed on the example of chlorophyllin 1% solution in ethyl alcohol at the wavelength of 417.5 nm, that to our knowledge has not yet been done.

Chlorophyllin is the semi-synthetic mixture of water-soluble sodium copper salts derived from chlorophyll [24] with the thermal origin of optical nonlinearity. Two main compounds of commercially synthesized chlorophyllin usually have a basic ring structure similar to chlorophyll (with only three differences in side chains and a changed central atom), but without a long phytol tail. Chlorophyllin was chosen as the nonlinear medium for this study because its nonlinear refractive index has not been previously considered in the literature, to the best of our knowledge. At the same time chlorophyllin should has strong optical nonlinearity. Such assumption can be based on two facts. Firstly, there is a number of the works [25–30], where tea solutions in water and alcohol were investigated. As it was shown in these studies, the high nonlinearity of thermal origin is caused by chlorophyll containing in tea leaves. Secondly, chlorophyllin has mainly almost the same shape of spectral absorbance [31] as chlorophylls *a and b* [32] in tea leaves (one peak around 650 nm and another next to 440 nm). That is why it also should possess similar high nonlinear properties in the spectral range of high chlorophyll absorbance. It should also be taken into account that chlorophyll and chlorophyllin have a number of useful applications. In particular, they possess antimutagenic and anticancirogenic effects [33–40], and also they can be used in photodynamic therapy [41,42]. We believe that the study of this easily affordable on the market chlorophyll derivative can complement the previous studies of tea solutions.

The paper is structured as follows: Section 2 describes two techniques used in this work to estimate the nonlinear refractive index of chlorophyllin solution in alcohol, as well as the approximations required for the proper applicability of these methods (subsection 2.1). First of them is the off-axis digital PI approach, which theoretical description, experimental set-up and the details about processing procedures are given in subsection 2.2. Second indirect approach chosen as a reference is based on the thermal equation analytical expression, which is derived for the same number of approximations. This technique is described in subsection 2.3. Section 3 presents the obtained results, and it is demonstrated that they have a quantitative close correlation with the reference approach, which confirms the validity of the data. The discussion of the obtained mismatch between experimental measurements and calculations based on the reference approach are given.

2. Methods

2.1. Admitted approximations

In general case, optical interferometry methods (including the mentioned above PI) can be used for the optical nonlinear properties investigation of the wide range of optical media with the varying type of optical nonlinearity. However, when the medium under investigation has the thermal mechanism of nonlinearity, additional requirements and experimental conditions should be fullfilled in order to obtain reliable data using interferometric approaches [43]. In particular, the nonlinear mechanism of chlorophyllin solution investigated in our work at the wavelength of 417.5 nm is primarily thermal. Consequently, in order to properly use the interferomtric method PI to study the nonlinear properties of this medium one should meet several additional requirement for the validity of the Kerr approximation.

These approximations and requirements for experimental measurements by the interferometric method PI are identical to those for the indirect method also used in our work. The indirect approach is based on the approximate expression (2) derived exclusively for the nonlinear media with the thermal origin of nonlinearity. The mentioned analytical equation was obtained in the work [23] from a general thermoconductivity equation on the basis of the mentioned above assumptions. The basis of this expression is a simple physical model describing a refractive index distribution in the course of Gaussian beam propagation in an absorbing medium in the absence of convection. This physical model allows one to calculate the value of refractive index change Δn, induced in the thermal nonlinear medium in the center of a Gaussian beam from its thermooptical parameters.

Therefore, for the medium under investigation with the thermal origin of nonlinearity a measurement process should be carried out when the following conditions and approximations are fulfilled: (i) all measurements should be conducted only when a steady-state temperature distribution takes place, (ii) the intensity of laser radiation should be small enough in order to neglect convection processes, (iii) optical radiation should has the Gaussian beam intensity distribution at the entrance of the nonlinear medium, (iv) any changes of the beam shape and size in the volume of the nonlinear medium (for example, due to the diffraction of light or nonlinear processes) should be vanishingly small. When experimental conditions comply with the requirements described above, the effective nonlinear refractive index can be determined by means of the digital holography as it will be described below.

2.2. Photothermal digital interferometry

2.2.1. Theory

First method used in this work is the Photothermal Interferometry implemented through the off-axis digital holography technique. In order to determine the value of the nonlinear refraction index n_2 within an interferometric approach, it is necessary to perform the reconstruction of a spatial phase distribution at the output plane of nonlinear medium using a holographic optical setup. Meanwhile, there are many interferometric techniques which can be used for this purpose. Choice of some or other optical method for the spatial phase distribution reconstruction depends primarily on the nonlinearity mechanism of the studied medium. For example, a similar approach was applied earlier in the work [44], where a pure water nonlinear refractive index was determined by a single-shot supercontinuum spectral interferometry method with femtosecond laser radiation at the wavelengths of 417 nm and 815 nm (the electronic-vibrational mechanism of nonlinearity). A simple formula approximately