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INTERFEROMETRIC METHOD FOR THE STUDY OF SPATIAL PHASE MODULATION INDUCED BY LIGHT IN DYE-DOPED DNA COMPLEXES

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Abstract. An interferometric pump-probe method for the investigation of the spatial phase modulation induced by light in nonlinear optical samples is presented. The optical phase of a probe beam passing through the sample is modified by the refractive index change induced by a pump beam. Consequently, the fringe pattern obtained at the output of the interferometer is modified. A Fourier transform algorithm for the direct spatial reconstruction of the optical phase from a single interference pattern is implemented. Using this method, the spatial distribution and the magnitude of the phase change induced by light in a dye-doped DNA complex are determined. These results are important for devices based on all-optical phase modulation.

Key words: interferometric method, spatial phase modulation, nonlinear refractive index, dye-doped DNA, Rh610 dye.

1. INTRODUCTION

The refractive index of a nonlinear optical material can be modified when it is illuminated by an intense light beam. This change of the refractive index can modulate the optical phase of the excitation beam itself or of other light beam passing through the considered nonlinear material. The nonlinear refractive response of the material is characterized by the nonlinear refractive coefficients, which can be experimentally determined by measuring the magnitude of the light induced phase change and by taking into consideration a certain mechanism of the optical nonlinearity. Several methods for deriving the nonlinear refractive index from the phase modulation have been proposed. In wave mixing method [1–4], two laser beams are generating a refractive index grating inside the investigated material. From the diffraction efficiency of this grating, monitored by a probe beam, the magnitude and the temporal evolution of the nonlinear phase modulation can be determined, but not its sign. In single beam methods, as Z-Scan and its derivative, I-Scan [5–12], the same laser beam is used to excite and to probe the optical nonlinearity in the investigated material. The conventional Z-scan methods provide the magnitude and the sign of the nonlinear phase change, but not its temporal evolution. Interferometric methods for the measurement of the nonlinear phase change have been also proposed [13–17].

In this paper, we present a modified experimental configuration of a single shot pump-probe interferometric method recently introduced [18] and use it for the measurement of the magnitude, sign and transversal spatial distribution of the phase modulation induced by light in deoxyribonucleic acid (DNA) – cetyltrimethyl-ammonium chloride (CTMA) – Rhodamine 610 (Rh610) complex in butanol. DNA-based materials are environmentally friendly, biodegradable, originating from renewable resources (waste of food processing industry) and have a high potential for applications in organic photonics and organic electronics.

2. THE INTERFEROMETRIC METHOD FOR THE STUDY OF LIGHT-INDUCED PHASE MODULATION

The spatial phase change induced by light in a nonlinear sample is investigated using the interferometric setup described in the Fig. 1, based on a Mach-Zehnder interferometer (MZI). This setup is a modified configuration of the one described in [18].

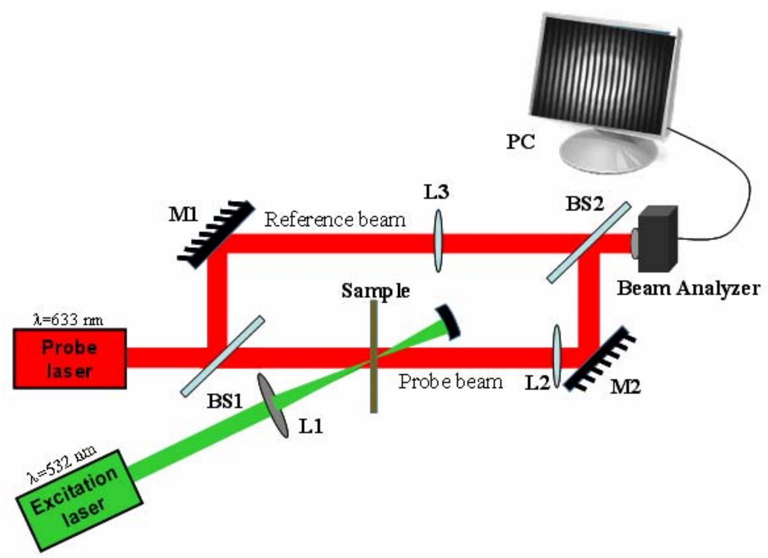


Fig. 1 – The interferometric setup for the study of light-induced phase modulation.

The nonlinear optical sample is placed in one arm of MZI. It is excited by a focused pump laser beam at a wavelength (λ) in the sensitivity range of the sample (in our case, λ = 532 nm, the second harmonic of a c.w. Nd:YAG laser) that locally changes its refractive index. The light source in the MZI is a probe laser with the transversal size of the beam at the sample plane larger than the one of the focused excitation beam. The wavelength of the weak probe beam is outside of the sensitivity range of the sample, so it is not changing its refractive index. In our study, a He-Ne laser (λ = 633 nm) is used as probe laser. The probe beam interferes with the reference beam on the photosensitive array of a Beam Analyzer (camera), producing a fringe pattern. This pattern stores the information about the phase difference between the two interfering beams.

The lens L2, placed in the interferometer between the sample and the camera, at distances equal to twice of its focal length, images the sample on the camera sensor with unitary magnification. Other desired magnifications can be obtained adjusting the position (and the focal length, if necessary) of the lens L2 relative to object and image planes, respectively. The lens L3 is placed in the other arm of the interferometer at the same distance to the camera as L2, in order to ensure the same curvature of the interfering beams wavefronts.

MZI is aligned so that in the absence of the excitation light (pump beam OFF), the fringe pattern consists in linear and parallel fringes (as shown as example in Fig. 2a). In this case, the phase difference $\Delta\Phi_{OFF}(x, y)$ is equal to the phase difference $\Delta\Phi_{tilt}(x, y)$ due to the angle between the interfering beams only:

$$\Delta\Phi_{OFF}(x,y) = \Delta\Phi_{tilt}(x,y). \tag{1}$$

When the sample is excited (pump beam ON), the optical phase of the probe beam is locally modified by the refractive index change induced by the pump beam and, consequently, it locally modifies the obtained interference fringe pattern (as shown as example in Fig. 2b). The phase difference $\Delta\Phi_{ON}(x, y)$, corresponding to this case, is:

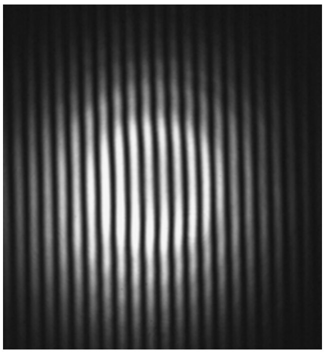
$$\Delta\Phi_{ON}(x,y) = \Delta\Phi_{tilt}(x,y) + \Delta\Phi_{NL}(x,y), \tag{2}$$

where $\Delta\Phi_{NL}(x, y)$ is the phase change induced by the excitation light in the nonlinear sample, directly related to the light-induced change of the refractive index, $\Delta n(x, y)$.

The phase differences $\Delta\Phi_{OFF}(x, y)$ and $\Delta\Phi_{ON}(x, y)$, respectively, are determined by processing the recorded interferograms using an algorithm based on Fourier transforms, implemented by us, for the direct spatial reconstruction of the optical phase (DSROP) from a single interference pattern [19–22].

$$\Delta\Phi_{OFF}(x,y)=\Delta\Phi_{tilt}(x,y)$$

(a)



$$\Delta\Phi_{ON}(x,y)=\Delta\Phi_{tilt}(x,y)+\Delta\Phi_{NL}(x,y)$$

(b)

Fig. 2 – The fringe patterns with the pump beam OFF (a) and ON (b), respectively, and the corresponding phase differences.

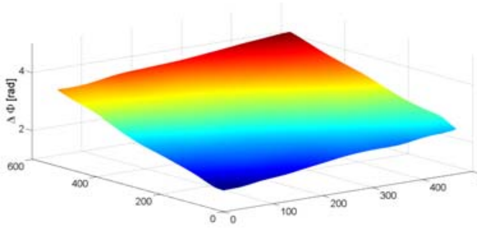
We will briefly present the main steps of the DSROP algorithm. After computing the Fourier distribution of the analyzed fringe pattern, one of the two first orders of the Fourier spectrum is selected and filtered in the Fourier plane. For a correct band-pass filtering of the desired term, the main spatial frequency in the fringe pattern (adjustable by the angle between the two interfering beams in the interferometer) must be sufficiently large to avoid the overlapping of different Fourier terms. By computing the inverse Fourier transform of the filtered Fourier term, the phase distribution can be obtained directly, wrapped in a 2π interval. Using an unwrapping algorithm, the continuous phase distribution, without 2π phase jumps, is determined from the wrapped phase distribution. The implemented DSROP algorithm is described in detail in [18].

The distribution of the light-induced phase change, $\Delta\Phi_{NL}(x, y)$, in the investigated sample is computed by subtracting from the phase change when the sample is optically excited, $\Delta\Phi_{ON}(x, y)$, the phase change due to the tilt only, $\Delta\Phi_{OFF}(x, y)$:

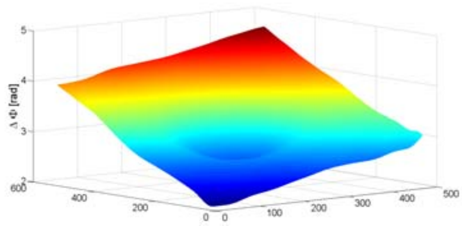
$$\Delta\Phi_{NL}(x,y)=\Delta\Phi_{ON}(x,y)-\Delta\Phi_{OFF}(x,y). \tag{3}$$

The phase distributions obtained from the fringe patterns shown in the Fig. 2, using the DSROP method previously described, are shown in the Fig. 3a,b. In the Fig. 3c is shown the light-induced phase modulation determined using Eq. (3).

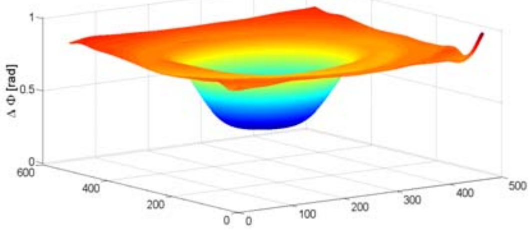
From the light-induced phase change determined as shown above, the light-induced change of the refractive index, Δn, can be determined. Considering a certain mechanism of the optical nonlinearity, the nonlinear refractive coefficients of the investigated material can be derived.



(a)
$\Delta\Phi_{OFF}(x, y)$



(b)
$\Delta\Phi_{ON}(x, y)$



(c)
$\Delta\Phi_{NL}(x, y)$

Fig. 3 – The phase distributions obtained from the fringe patterns without (a) and with (b) light excitation, and the light-induced phase change (c), computed by subtracting (a) from (b).

We exemplify below the relation between the light-induced changes of the optical phase and of the refractive index, for the case of the third-order optical nonlinearity excited in the sample. In this case Δn is proportional to the intensity of the pump beam, $I_{\text{pump}}(x, y)$:

$$\Delta n(x,y) = n_2 \cdot I_{pump}(x,y), \tag{4}$$

n_2 being the nonlinear refractive index.

In samples with negligible absorption of the pump light, the change of the refractive index induced by the pump beam is uniform along the propagation path of this beam through the sample, and the light-induced phase change accumulated by the probe beam until the exit from the sample of thickness L is:

$$\Delta\Phi_{NL}(x,y,z=L) = k \cdot \Delta n(x,y) \cdot L, \tag{5}$$

where $k = 2\pi/\lambda_{\text{probe}}$ is the wave number of the probe beam. From Eqs. (4) and (5), n_2 is related to the light-induced phase change by:

$$n_2 = \frac{\Delta\Phi_{NL}(x,y,z=L)}{k \cdot I_{pump}(x,y) \cdot L}. \tag{6}$$

In samples with non-negligible absorption, α_0, the exponential decrease (Lambert-Beer law) of the intensity of the pump beam along its path through the sample (*z* coordinate), given by:

$$I_{pump}\left(x,y,z\right)=I\left(x,y,z=0\right)\cdot \mathrm{e}^{-\alpha_0\cdot z}, \qquad (7)$$

must be taken into account. In this case, the light-induced phase change accumulated by the probe beam until the exit from the sample is:

$$\Delta\Phi_{NL}\left(x,y,z=L\right)=k\cdot\int_0^L \Delta n\left(x,y,z\right)\cdot \mathrm{dz}=k\cdot n_2\cdot I(x,y,z=0)\cdot L_{eff}, \qquad (8)$$

$$L_{eff}=\int_0^L \mathrm{e}^{-\alpha_0\cdot z}\mathrm{dz}=\left(1-\mathrm{e}^{-\alpha_0\cdot L}\right)/\alpha_0, \qquad (9)$$

L_{eff} being the effective length of the sample. The nonlinear refractive index n_2 in samples with non-negligible absorption is:

$$n_2=\frac{\Delta\Phi_{NL}\left(x,y,z=L\right)}{k\cdot I_{pump}\left(x,y,z=0\right)\cdot L_{eff}}. \qquad (10)$$

3. INVESTIGATION OF THE LIGHT-INDUCED SPATIAL PHASE MODULATION IN THE COMPLEX DNA-CTMA-Rh610 IN BUTANOL USING THE INTERFEROMETRIC METHOD

The investigated sample is the complex DNA-CTMA in butanol (30 g/l) doped with Rh610 (10% wt). The concentration represents the percentage of the dye mass with respect to the matrix (DNA-CTMA) dry mass. Details about sample preparation can be found in [23], in which the experimental demonstration of lasing in this complex is reported. The sample is placed in a thin quartz cuvette (L = 0.5 mm thickness), with optical quality windows.

The absorption spectrum of the sample is shown in Fig. 4. The wavelength of the pump light (marked with green in the Fig. 4) is near the absorption peak of the sample (λ = 542.6 nm), the pump light practically being completely absorbed in our sample.

The Gaussian pump beam is focused on the sample to a spot with the diameter: D_{pump} = 400 μm, approximately four times smaller than the probe beam spot. The peak intensity of the pump beam was tuned in the range I = (0.27÷22.61) W/cm^2 by tuning its peak power from P = 0.17 mW to P = 14.2 mW.

A selection of the locally distorted fringe patterns, at several intensities of the pump beam, and of the corresponding light-induced phase modulations, $\Delta\Phi_{NL}$, extracted from them using the implemented DSROP method are shown in the

Fig. 5a–g. The magnitude of the phase modulation is determined in the center of the phase-modulated region, excited by the peak of the Gaussian pump beam.

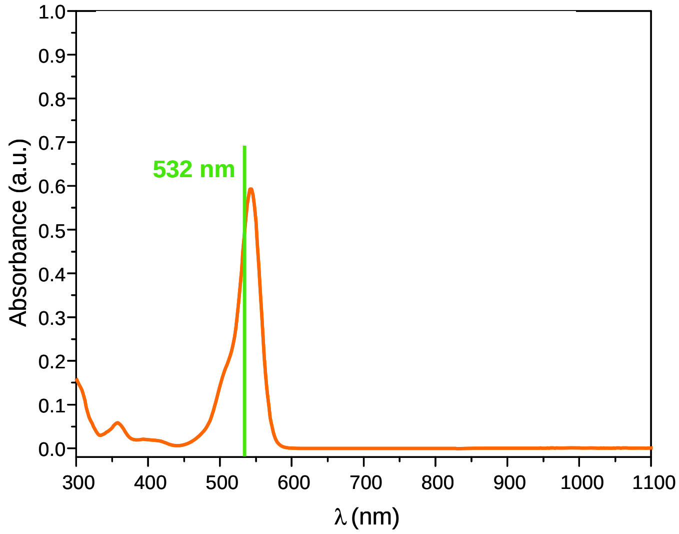
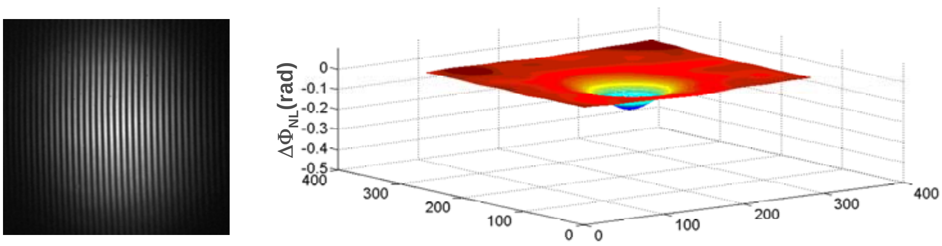
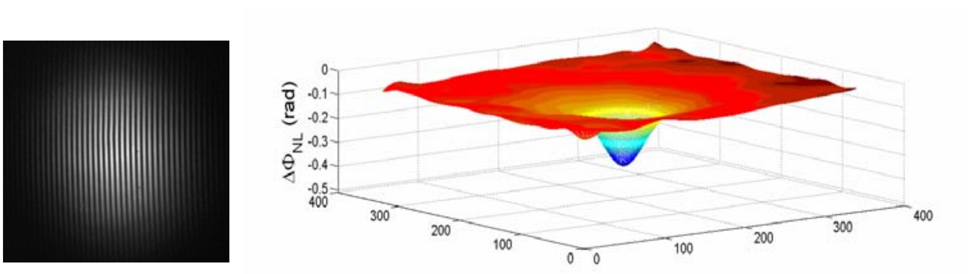


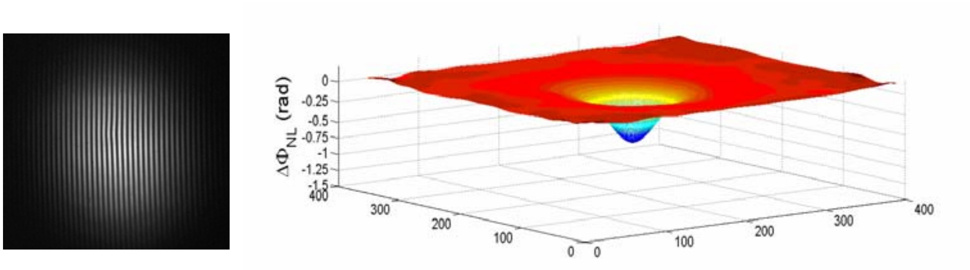
Fig. 4 – The absorption spectrum of the complex DNA-CTMA-Rh610 in butanol.



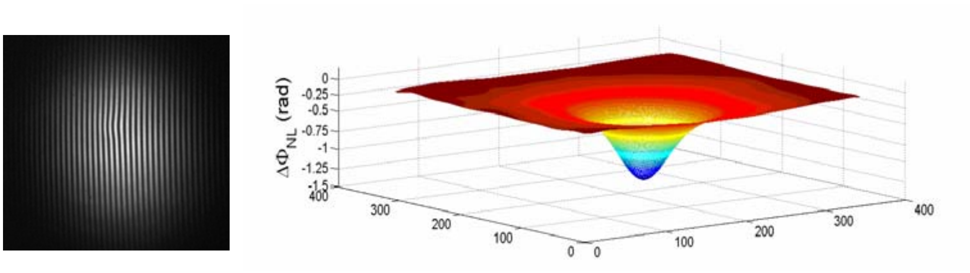
(a) $I = 0.27$ W/cm^2, $\Delta\Phi_{NL} = -0.13$ rad



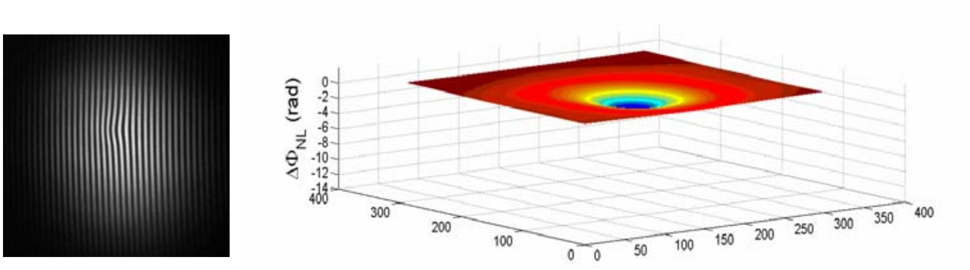
(b) $I = 0.66$ W/cm^2, $\Delta\Phi_{NL} = -0.30$ rad



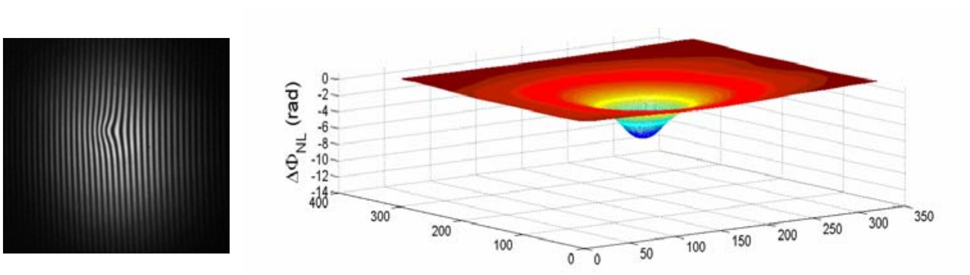
(c) I = 1.24 W/cm^2, $\Delta\Phi_{NL}$= –0.67 rad



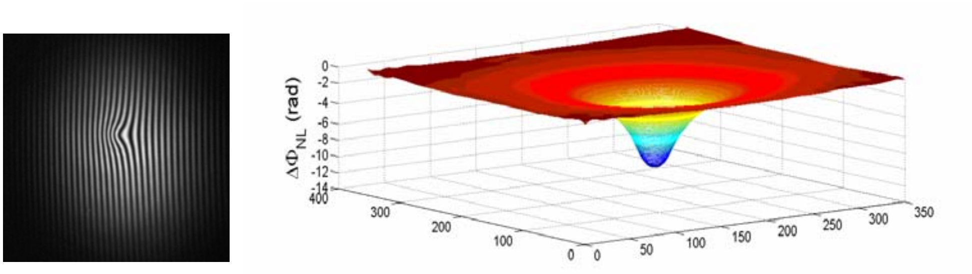
(d) I = 2.90 W/cm^2, $\Delta\Phi_{NL}$= –1.24 rad



(e) I = 5.96 W/cm^2, $\Delta\Phi_{NL}$= –2.44 rad



(f) I = 13.70 W/cm^2, $\Delta\Phi_{NL}$= –6.15 rad



(g) $I = 22.61$ W/cm^2, $\Delta\Phi_{NL} = -10.58$ rad

Fig. 5 – The distorted fringe patterns and the computed light-induced phase modulations $\Delta\Phi_{NL}$, at several intensities of the pump beam. The vertical scale is different: $|\Delta\Phi_{NL}| < 0.5$ rad (a, b); $|\Delta\Phi_{NL}| < 1.5$ rad (c, d); $|\Delta\Phi_{NL}| < 15$ rad (e, f, g).

The dependence of the phase change magnitude on the pump intensity, derived from all experimental data, is shown in Fig. 6.

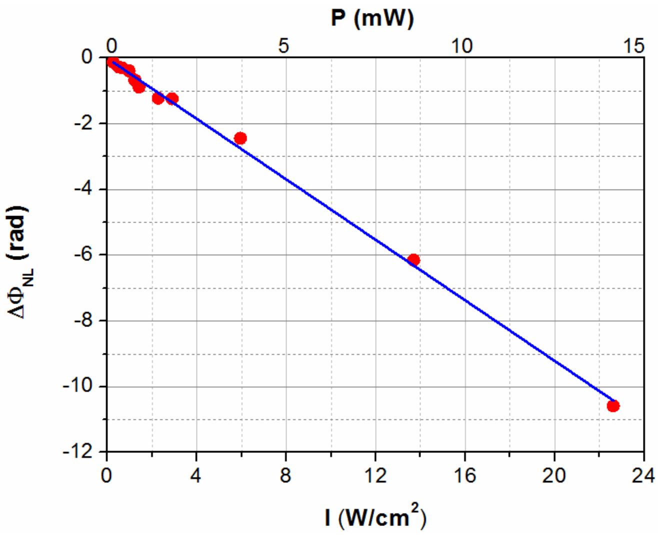


Fig. 6 – The dependence of the phase change induced by light in DNA-CTMA-Rh610 sample on the pump intensity.

In the range $I = (0.27 \div 22.61)$ W/cm^2 of the pump intensities used in our experiments, the dependence of $\Delta\Phi_{NL}$ on I is well fit by a linear one:

$$\Delta\Phi_{NL}\left[\text{rad}\right] = -0.46\left[\frac{\text{rad}}{\text{W}/\text{cm}^2}\right]\cdot I\left[\text{W}/\text{cm}^2\right]. \quad (11)$$

The negative sign of the light-induced phase modulation, as seen in Fig. 5, corresponds to a divergent lens induced by light in the sample (defocusing nonlinearity).

4. CONCLUSIONS

A method for the investigation of the spatial phase modulation induced by light in nonlinear materials, based on an interferometric pump-probe configuration, has been introduced. A Fourier transform algorithm for the direct spatial reconstruction of the optical phase from a single interference pattern has been implemented. The magnitude, sign and transversal spatial distribution of the phase modulation induced by light at 532 nm wavelength in DNA-CTMA-Rh610 complex in butanol have been determined using this method.

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REFERENCES

1. H. J. Eichler, P. Günter, D. W. Pohl, *Laser induced dynamic Gratings*, Springer Verlag, Berlin, Heidelberg, New York, Tokio, 1986.
2. V. I. Vlad, E. Fazio, M. J. Damzen, A. Petris, Dynamic Waveguides and Gratings in Photorefractive Crystals, in *Photo-Excited Processes, Diagnostics and Applications*, A. Peled, Ed., Springer US, 2004, 57–100.
3. A. Petris, M. J. Damzen, V.I. Vlad, Opt. Commun. **176**, 223 (2000).
4. A. Petris, E. Fazio, T. Bazaru, V. I. Vlad, Rom. Rep. Phys. **53**, 727 (2001).
5. M. Sheik-Bahae, A. A. Said, E. W. Van Stryland, Opt. Lett. **14**, 955 (1989).
6. M. Sheik-Bahae, A. Said, T. H. Wei, D. J. Hagan, E. W. Van Stryland, IEEE J. Quantum Electron. **26**, 760 (1990).
7. T. Xia, D.J. Hagan, M. Sheik-Bahae, E.W. Van Stryland, Opt. Lett. **19**, 317 (1994).
8. B. Taheri, H. Liu, B. Jassemnejad, D. Appling, R.C. Powell, J.J. Song, Appl. Phys. Lett. **68**, 1317 (1996).
9. I. Dancus, V. I. Vlad, A. Petris, I. Rau, F. Kajzar, A. Meghea, A. Tane, Proc. SPIE **8882**, 88820D (2013).
10. I. Dancus, V. I. Vlad, A. Petris, T. Bazaru Rujoiu, I. Rau, F. Kajzar, A. Meghea, A. Tane, Rom. Rep. Phys. **65**, 966 (2013).
11. T. Bazaru Rujoiu, V. I. Vlad, A. Petris, Rom. Rep. Phys. **65**, 954 (2013).
12. T. Bazaru Rujoiu, A. Petris, V. I. Vlad, Proc. SPIE **8882**, 88820C (2013).
13. G. Boudebs, M. Chis, X. Nguyen Phu, J. Opt. Soc. Am B **18**, 623 (2001).
14. V. Besse, C. Cassagne, G. Boudebs, Journal of Modern Optics **59**, 887 (2012).
15. M. J. Moran, C. Y. She, R. L. Carman, IEEE J. Quantum Electron. **QE-ll** 259 (1975).
16. M. J. Weber, D. Milam, W. L. Smith, Opt. Eng. **17**, 463 (1978).
17. D. De Feo, S. De Nicola, P. Ferraro, P. Maddalena, G. Pierattini, Pure Appl. Opt. **7**, 1301 (1998).
18. I. Dancus, S. T. Popescu, A. Petris, Opt. Express **21**, 31303 (2013).
19. M. Takeda, H. Ina, S. Kobayashi, J. Opt. Soc. Am. **72**, 156 (1982).
20. V. I. Vlad, D. Malacara, Prog. Optics **33**, 261 (1994).
21. W. W. Macy, Appl. Optics **22**, 3898 (1983).
22. V. I. Vlad, D. Malacara, A. Petris, Opt. Eng. **35**, 1383 (1996).
23. T. Bazaru Rujoiu, A. Petris, V.I. Vlad, I. Rau, A.M. Manea, F. Kajzar, Phys. Chem. Chem. Phys. **17**, 13104 (2015).