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Transformation of Topological Structure of Optical Vortices upon Frequency Non-Degenerate Four-Wave Mixing

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The method for topological structure transformation of optical vortices with simultaneous frequency conversion in a medium with resonant and thermal nonlinearities was theoretically introduced on the basis of holographic principles for optical image processing. Frequency non-degenerate four-wave mixing of singular light beams was realized experimentally when recording and reading of nonlinear dynamic holograms in organic dye solutions.

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1. Introduction

Optical vortices, which are the subject of permanent interest in modern optics due to numerous potential applications, have a screw surface in their wave front structure with a point of singularity at which the field amplitude is equal to zero and the phase is undefined [1]. Nowdays, the main directions of investigation in this area are related to the study of the methods of generation of the singular light beams and transformation of their topological structure [2, 3]. Thus, recent experimental studies of four-wave mixing in atomic vapors and colloid crystals [4] made it possible to develop a method of direct detection of the phase conjugation from inversion of topological charge in an optical vortex. The possibility of implementing optical computations with optical vortices has been demonstrated experimentally on the base of the frequency non-degenerate four-wave mixing in Rb-vapors [5]. The method of second harmonic generation was used for realization of frequency doubling of optical vortices [6].

Among the wave front conversion methods for three-dimensional images characterized by a complex wave front, of particular interest are those based on nonlinear dynamic hologram recording [7]. The use of resonant media including the solutions of complex organic compounds looks promising for realization of the process over wide temporal and spectral ranges. The formation of dynamic holograms in such media is caused by changes in the refractive index and absorption coefficient when the molecules are activated to higher energy states and also by thermal nonlinearity due to the medium heating.

The use of light waves with different frequencies for recording and reading of the three-dimensional dynamic diffraction structures in nonlinear media provides an opportunity to perform real-time frequency conversion of coherent images. In the well-known schemes used for recording and reading of dynamic holograms, the conditions of Bragg diffraction are usually satisfied by a change the direction of a reading wave [8].

The main goal of this paper is to theoretically justify and experimentally realize the method for transformation of topological structure of the singular light beams with simultaneous frequency conversion when using holographic principles of optical image processing. It is shown that on the base of frequency non-degenerate four-wave mixing one can perform both inversion of topological charge of the vortex light beam and its frequency conversion

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from IR to visible range of optical spectrum. The work is organized as follows: Section 2 presents the theoretical model of the vortex light beam interaction in nonlinear medium, while the Section 3 is devoted to experimental investigation of the process of vortex light beams transformation upon realization of frequency non-degenerate four-mixing in organic dye solution.

2. Theoretical model

Let us consider the scheme when a dynamic hologram is recorded by the signal, $E_S = A_S \exp\left[i\left(\vec{k}_S \cdot \vec{r} - \omega t + \varphi_S\right)\right]$, and reference, $E_1 = A_1 \exp\left[i\left(\vec{k}_1 \cdot \vec{r} - \omega t + \varphi_1\right)\right]$, waves. Reading is performed by the wave $E_2 = A_2 \exp\left[i\left(\vec{k}_2 \cdot \vec{r} - 2\omega t + \varphi_2\right)\right]$ at the doubled frequency (see Fig.1). In the process of the light waves interaction in nonlinear medium with a susceptibility $\chi^{(3)}$, the polarization responsible for the generation of the wave E_D at the doubled frequency 2ω, is written as $P \propto E_1 E_S^* E_2$. Then, the phase-matching conditions $\vec{k}_1 + \vec{k}_2 = \vec{k}_S + \vec{k}_D$ corresponds to a decreased angle between the diffracted and reading waves as compared to the angle between the beams involved in hologram recording (Fig.1).

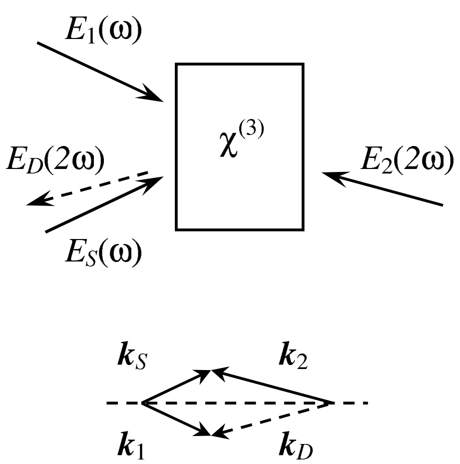

FIG. 1. The scheme of non-degenerate four-wave mixing and the diagram of the associated wave vectors

For a theoretical description of the above-described interaction scheme we assume that the medium absorbs radiation at frequency ω and is transparent at the doubled frequency 2ω. Then, the wave E_D is formed due to diffraction of the reading wave E_2 from the thermal dynamic grating written by the signal and reference waves. Further analysis will be performed for a two-level model of the resonant medium taking into account thermalization of energy in nonlinear medium due to nonradiative transitions in resonant channel $S_0 - S_1$. Based on a system of kinetic equations for the level population and on Kramers-Kronig (dispersion) relations for the refractive index and absorption coefficient, the total nonlinear (resonance and thermal) susceptibility of the medium for the system under consideration can be given in the following form [7]:

$$\chi_{nl}(\omega) = \frac{n_0 \kappa_0}{2\pi}\left(\frac{\hat{\Theta}_{12}}{B_{12}} - \frac{\hat{\alpha} I}{1 + \alpha I}\right) \qquad (1)$$

$$\chi_{nl}(2\omega) = \frac{n_0 \kappa_0}{2\pi}\left(\frac{a_T I}{1 + \alpha I}\right) \qquad (2)$$

where $\hat{\alpha} = a + i\alpha = (\hat{\Theta}_{12} + \hat{\Theta}_{21})/vP_{21} - a_T$, $a_T = \sigma_T\left(1 - \mu_{21}\right)$, $\alpha = \left(B_{12} + B_{21}\right)/vP_{21}$. The parameter α determines a saturation intensity of the resonant channel $S_0 - S_1$ $\left(I_{SAT} = \alpha^{-1}\right)$, $I = I_1 + I_S + 2\sqrt{I_1 I_S}\cos\left[\left(\vec{k}_1 - \vec{k}_S\right) \cdot \vec{r} + \varphi_1 - \varphi_S\right]$. Here, B_{12} and B_{21} are the Einstein coefficients for the induced transitions in $S_0 - S_1$ channel, v is the light velocity in the medium, κ_0 is the initial extinction coefficient, n_0 is the refractive index, P_{21} is the total probability of spontaneous and nonradiative transitions in channel $S_0 - S_1$, $\hat{\Theta}_{ij}(\omega) = \Theta_{ij}(\omega) + iB_{ij}(\omega)$ (coefficients $\Theta_{ij}(\omega)$ are related by Kramers-Kronig relations to Einstein coefficients $B_{ij}(\omega)$), $\sigma_T = 2\omega\left(\partial n/\partial T\right)\tau/cC_\rho$, τ is an interaction duration, C_ρ – heat capacity for the unit volume, $(\partial n/\partial T)$ - thermooptical coefficient, μ_{21} – luminescence quantum yield in channel $S_0 - S_1$.

For the considered case of co-propagating signal E_S and reference E_1 waves (Fig.1), a set of coupled-mode equations describing the process of frequency non-degenerate four-wave mixing with due regard for the transverse structure of recording, reading and diffracted light beams has the following form:

$$\left(\frac{\partial}{\partial z} \mp \gamma_{1,S}\frac{\partial}{\partial x} + \frac{\Delta_\perp}{2ik_{1,S}}\right) E_{1,S} = \frac{i2\pi\omega}{cn_0}\left[\chi_0(\omega)E_{1,S} + \chi_{\pm 1}E_{S,1}\right] \quad (3)$$

$$\left(\frac{\partial}{\partial z} \pm \gamma_{2,D}\frac{\partial}{\partial x} + \frac{\Delta_\perp}{2ik_{2,D}}\right) E_{2,D} = -\frac{i4\pi\omega}{cn_0}\left[\chi_0(2\omega)E_{2,D} + \chi_{\mp 1}E_{D,2}\right] \quad (4)$$

where $\chi_m = (1/2\pi)\int_{-\pi}^{\pi}\chi(\zeta)\exp[-im\zeta]d\zeta$ are the Fourier-series expansion components of the medium nonlinear susceptibility χ in the grating harmonics $\zeta = \left(\vec{k}_1 - \vec{k}_S\right)\cdot\vec{r}$ determined by the optical and spectroscopic characteristics of the resonance transition, as well as by the frequency and intensity of the interacting waves. In equations (3) - (4), $\gamma_{1,2,S,D}$ are the angles between the z axis and the wave vectors $\vec{k}_1, \vec{k}_2, \vec{k}_S, \vec{k}_D$, respectively; $k_{1,S} = \omega n_0/c$, $k_{2,D} = 2\omega n_0/c$ are the wave numbers; and $\Delta_\perp = \partial^2/\partial x^2 + \partial^2/\partial y^2$ is the transverse Laplacian.

Using the explicit form of the expressions for the Fourier-components of the nonlinear susceptibility, one can reduce the set of Eqs. (3) and (4) to the form:

$$\left(\frac{\partial}{\partial z} \mp \gamma_{1,S}\frac{\partial}{\partial x} + \frac{\Delta_\perp}{2ik_{1,S}}\right) E_{1,S} = i\frac{k_0}{2} f_{1,S}E_{1,S} \quad (5)$$

$$\left(\frac{\partial}{\partial z} \pm \gamma_{2,D}\frac{\partial}{\partial x} + \frac{\Delta_\perp}{2ik_{2,D}}\right) E_{2,D} = -ik_0\left[\psi E_{2,D} + \phi E_{D,2}\right] \quad (6)$$

where $k_0 = 2\omega\kappa_0/c$ is the linear absorption coefficient,

$$f_{1,S} = \frac{\Theta_{12}}{B_{12}} - \frac{a}{\alpha} + \frac{\hat{\alpha}}{\alpha A} - \frac{2\alpha I_{S,1}}{A\left[1 + \alpha\left(I_1 + I_S\right) + A\right]},$$

$$\psi = \frac{a_T}{\alpha}\left(1 - \frac{1}{A}\right),$$

$$\phi = \frac{a_T}{\alpha}\frac{2\alpha\sqrt{I_1 I_S}}{A\left[1 + \alpha\left(I_1 + I_S\right) + A\right]}\exp\mp i\left(\varphi_1 - \varphi_S\right),$$

$$A = \left[1 + 2\alpha\left(I_1 + I_S\right) + \alpha^2\left(I_1 - I_S\right)^2\right]^{1/2},$$

and $I_{1,S} = cn_0\left|E_{1,S}\right|^2/8\pi$ are the intensities of the reference and signal waves.

The functions f_1 and f_S include both the modulation of the absorption coefficient and the refractive index due to self-transparency of the medium in the interference field of the reference and signal waves, ψ describes self-modulation, and ϕ characterizes a parametric coupling between the waves E_2 and E_D due to diffraction from spatial harmonics of the dynamic grating recorded in the medium by the waves E_1 and E_S [9].

In the following analysis, we suppose that the signal light beam contains a helical phase dislocation with the topological charge m, the simplest type of which can be mathematically described by a complex input light field E_S in the form:

$$E_S(z=0,\rho,\varphi) = A_S\left[(r - r_S)/r_{0S}\right]^{|m|} \times \exp\left[-\left(\left(r - r_S\right)/\sqrt{2}r_{0S}\right)^2 + im\varphi\right]$$

Here, ρ and φ are polar coordinates, parameter r_{0S} is characterized the width of light beam, $m = \pm 1, \pm 2, ...$ is the so-called topological charge of optical vortex. Let us notice that $m = 0$ corresponds to the case of Gaussian light beam.

The reference E_1 and reading E_2 beams have a plane wave front with the amplitudes:

$$E_1(z=0,\rho,\varphi) = A_1\exp\left[-\left(\left(r - r_1\right)/\sqrt{2}r_{01}\right)^2\right]$$

$$E_2(z=L,\rho,\varphi) = A_2\exp\left[-\left(\left(r - r_2\right)/\sqrt{2}r_{02}\right)^2\right]$$

and half-width $r_{01} = r_{02} = 3r_{0S}$. In numerical simulation, we suppose that recording light beams are crossed in the nonlinear medium at the angle $2\gamma_1 = 40 mrad$, and the initial distance between their centres at the boundary $z = 0$ is $r_1 - r_S = r_{0S}$, where $r_{0S} = 0.1cm$, the reference beam intensity is $\alpha I_0 = 1$, and signal to reference intensity ration is $I_{0S}/I_0 = 0.1$. The parameters of nonlinear medium are the following: the linear absorption coefficient of nonlinear medium is $k_0 = 1cm^{-1}$, the length of nonlinear layer is $L = 1cm$, refractive index is $n_0 = 1.36$, thermooptical coefficient $(\partial n/\partial T) = -4 \cdot 10^{-4}K^{-1}$, heat capacity for the unit volume $C_\rho = 2JK^{-1}cm^{-3}$, pulse duration $\tau = 20ns$, $\lambda = 1\mu m$, $\Delta\lambda = 100nm$ (λ and $\Delta\lambda$ are the center and half-width of the absorption band, respectively), $\mu_{21} = 0.01$. The medium was excited at the center of the absorption band, and the mirror-symmetric luminescence band was Stokes-shifted by $\Delta\lambda$.

The numerical modelling of Eqs. (5) and (6) was performed using absolutely stable two-step (three-layer) explicit method [10] that gives a possibility to calculate 2D spatial intensity and phase distribution of interacting light beams in the bulk of nonlinear medium. Let us notice that the geometry of interaction under consideration implies solution of the boundary-value problem with the boundary conditions specified at different boundaries of the nonlinear medium (the fields E_1 and E_S are determined at the boundary $z = 0$, while the field of reading wave E_2 is directed toward the boundary $z = L$). For this reason, the numerical simulation was performed in two steps. First, we calculated the profiles of the signal and reference beams in the bulk of the nonlinear medium [direct solution of Eqs. (5)], then we solved the total system of Eqs. (5) and (6) backward from the boundary $z = L$ to $z = 0$ and found spatial profiles of the reading and diffracted beams.

An analysis of the phase-matching conditions and Eq. (6) shows that in the process of four-wave mixing with the use of the plane reference and reading waves ($\varphi_1 + \varphi_2 = const$), the phase of the wave diffracted from dynamic grating is opposite to that of the signal wave ($\varphi_D = -\varphi_S$). Thus, when signal beam contains an m-order screw phase dislocation, the wave front of the diffracted beam must contain dislocation of the opposite sign ($-m$).

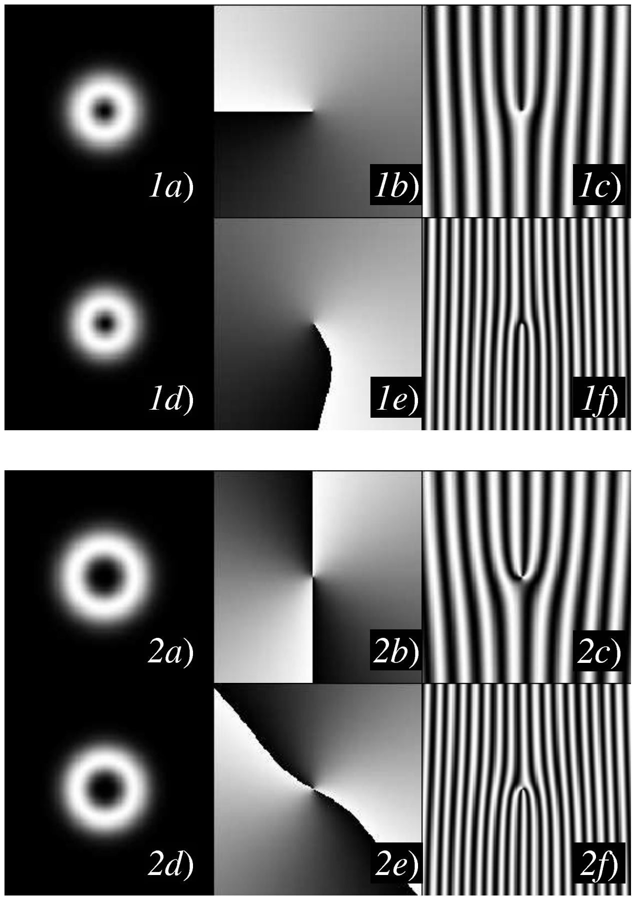

FIG. 2. Intensity (a, d), phase distribution (b, e), and interference patterns (c, f) for signal (a-c), and diffracted (d-f) light beams. $m = 1$ - (1a-1f), $m = 2$ - (2a-2f).

The results of numerical simulation of the system of Eqs. (5) and (6) are shown in Fig.2. As one can see, due to diffraction of the reading wave E_2 from the phase grating written by the reference beam E_1 and signal beam E_S, containing screw dislocation of the topological charge $m = 1$ (1a, 1b in Fig.2), a diffracted beam E_D is formed at doubled optical frequency 2ω (1d, 1e in Fig.2) whose wave front contains the dislocation of opposite sign ($m = -1$). This fact is also confirmed by the structure of of the signal and diffracted beams with the plane reference wave (compare 1c and 1f in Fig.2). In the same way, the transformation of singular light beams with higher orders of topological charge can be realized (see 2a-2f in Fig.2).

As one can see from Fig.2, the diffracted light beam at doubled optical frequency 2ω is characterized by the wave front distorted with respect to the signal beam. A phase shift $\Delta\varphi \approx \pi/2$ for the topological charge $m = 1$ arises due to predominant contribution in diffraction of the phase gratings at doubled optical frequency 2ω. In addition, the nonlinear variation of the refractive index results in an incomplete restoration of the wave front structure.

3. Experimental results

The transformation of topological structure of the singular light beams with simultaneous frequency conversion has been experimentally realized in ethanol solution of the polymethine dye 3274U by the diffraction of laser radiation from transmission dynamic hologram formed by co-propagating signal and reference waves.

The experimental setup is shown in Fig.3. The experiments were performed with an yttrium-aluminum garnet laser 1 (lasing wavelength $\lambda = 1.06\mu m$, laser beam divergence at half maximum $\theta_{0.5} \leq 2mrad$, pulse duration $\tau = 20ns$). A dynamic hologram was recorded at the fundamental frequency of laser 1 associated with the absorption band maximum of the polymethine dye 3274U in ethyl alcohol (saturation absorption intensity $I_{SAT} = 13MW/cm^2$ at the wavelength $\lambda = 1.06\mu m$). Holograms were reconstructed by the second harmonic radiation of the same laser ($\lambda = 0.532\mu m$), being practically unabsorbed by the dye solution. The spatially homogeneous portion of radiation was cut out by diaphragm 2. The signal E_S and reference E_1 waves were formed by beam splitter 3 and mirrors 5, 6. The intensity of reference and signal beam was $I_1 = 1MW/cm^2$, and $I_S = 0.1I_1$. The reading wave E_2 at a double frequency was directed at a small angle relative to reference wave E_1 with the help of mirror 4 to fulfill the phase-matching conditions for frequency non-degenerate four wave mixing. An angle ($\gamma \approx 30mrad$) between the propagation directions of the reference and signal beams and its transversal size ($r_0 = 250\mu m$, $r_{0S} = 75-150\mu m$) offered overlapping of the interacting waves along the full length of cell 7 containing the dye solution. Spatial profiles of the signal and diffracted light beams were photographed with a CCD-camera placed in respective arm of experimental setup.

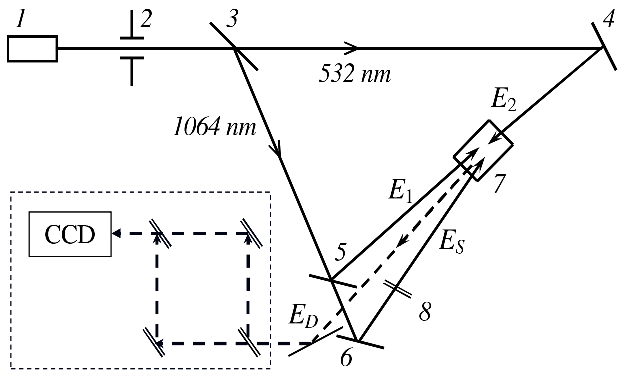

FIG. 3. Experimental setup for realization of frequency non-degenerate four-wave mixing with singular light beams

A computer-synthesized phase hologram 8 fixed in PMMA layers containing phenanthrenequinone [11] was used to obtain the signal beam with a screw dislocation in its wave front structure. The intensity level of reference and signal beams used in experiments was lower than irradiance destruction threshold level for transparency 8 ($11MW/cm^2$ for $\lambda = 1.06\mu m$, and pulse frequency $\nu = 10Hz$), determined by measurement of their diffraction efficiency in different excitation conditions [11]. The diffraction efficiency of transmission-type transparency 8 was $\xi = 0.08$ both for first ($m = 1$) and second ($m = 2$) order of the singular light beams.

Mach-Zender interferometer, which is shown in the insertion of Fig.3, was placed in an appropriated arm of experimental setup to obtain interference patterns for signal and diffracted light beams and to reveal the consequent topological structure of light beam. The used

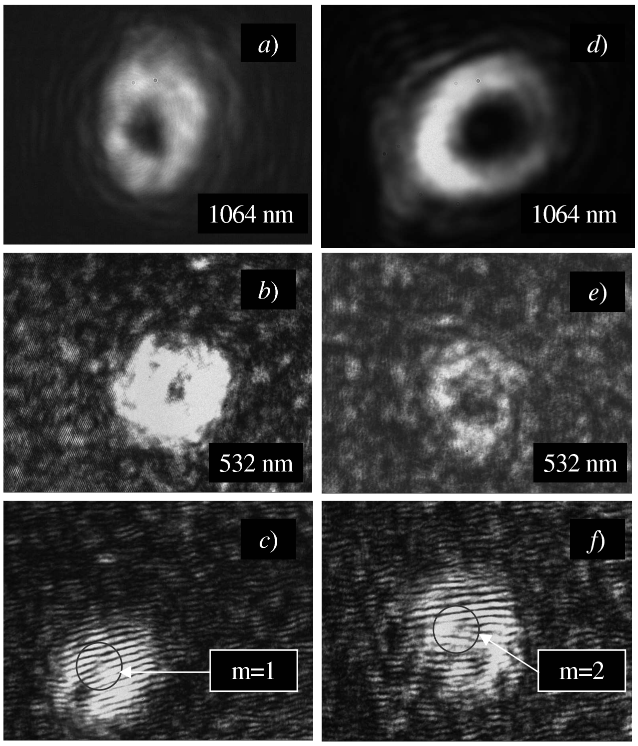

FIG. 4. Experimental measurements of spatial profiles of signal (a, d), diffracted (b, e) light beams, and interference pattern for diffracted beam (c, f). $m = 1$ - (a, b, c), $m = 2$ - (d, e, f).

configuration of Mach-Zender interferometer permits to obtain an interference pattern of slightly tilted light beams, and to reveal their topological structure. Let us notice, the contrast of interference patterns in the area of localization of optical vortices obtained with the used method is significantly higher in comparison with the work [4].

The results of experimental investigation of frequency non-degenerate four-wave mixing in ethanol solution of polymethine dye $3274U$ are shown in Fig.4. As one can see, due to diffraction of the reading wave E_2 from the phase grating written by the reference beam E_1 and signal beam E_S, containing a screw dislocation of the topological charge $m = 1$ (Fig.4a), the diffracted beam E_D with the same order of topological charge is formed at doubled optical frequency 2ω (Fig.4b). The interference pattern from Michelson interferometer (Fig.4c) is characterized by the presence of a single forked structure, which is a typical for optical vortices with $m = 1$. In the same way, the transformation of singular light beams with topological charge $m = 2$ was realized (Fig.4d-f).

4. Conclusions

Thus, the above theoretical analysis and numerical simulation show that the scheme of frequency non-degenerate four-wave mixing in the medium with resonant and thermal nonlinearities can be used to change the sign of the topological charge of singular light beams with simultaneous frequency conversion. The method for transformation of the topological charge of optical vortices with frequency conversion from IR to visible range of optical spectrum, which is based on non-degenerate four-wave mixing, was experimentally implemented with the use of ethanol solution of polymethine dye $3274U$ as a nonlinear medium.

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