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Multiwave Mixing of Singular Light Beams in Resonant Media

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Abstract—A method for transforming the topological structure of a singular light beam under multiwave mixing in the media with resonant nonlinearity is justified theoretically. The effect of self- and cross-modulation of the light waves under condition of nonlinear variation of the refractive index on stability of the optical vortices appearing in the multiwave mixing is studied.

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A permanent interest in singular optics is related to numerous potential applications of light beams with the wavefront dislocations. Nowadays, the main trends in this research are related to the study of the methods of generation and transformation of the singular light beams [1], solution of the problem of stability of the spatial and topological structure of optical vortices upon propagation through the linear and nonlinear media [2], application of optical vortices for implementation of optical computations [3], the study of the waveguide properties of the spatially localized optical vortices [4], and analysis of polarization structure of the singular light beams [5]. The methods for applying singular beams as "optical tweezers" for optical trapping and manipulation of single molecules and microparticles are being intensely developed [6].

In recent years, a considerable amount of attention has been paid to the problems of nonlinear interaction of optical vortices, as well as to the problems of controlling the topological charge and polarization structure of singular light fields. Development of methods of transformation of topological structure of the optical vortices, among which are the second harmonic generation [7] and three- and four-wave mixing [8], is of interest for applications related to optical computations. Recent experimental studies of the phase conjugation effect in atomic vapors and colloid crystals [9] made it possible to develop a method of direct detection of the phase conjugation from inversion of topological charge in a singular signal beam. The possibility of implementing optical computations with the use of topological charges of the optical vortices based on the method of the frequency-nondegenerate four-wave mixing has been demonstrated experimentally [10].

The development of nonlinear interferometric and holographic systems based on multiwave mixing in resonant media [11] has shown wide prospects of their use for transformation of the wavefront of optical vortices, for the interbeam information transfer, for implementation of the logical and mathematical operations, and for creating elements of adaptive optics.

The main goal of this paper is to theoretically justify the possibility and to study efficiency of transforming the topological structure of singular light beams when using nonlinear holographic methods of optical image processing. We present the results of theoretical analysis and numerical simulation of the processes of multiwave mixing of singular beams in resonant media.

The recording and reading of dynamic holographic gratings arising upon the interaction of light beams in resonant media under conditions for the saturation of light-induced changes in the absorption coefficient and (or) in the refractive index are characterized by certain specific features. Let the gratings be recorded by the signal (E_S) and reference (E_1) waves with the frequency ω close to that of the resonance transition of the nonlinear medium. For a counter-propagating reading wave E_2 with the same frequency ω, a nonlinear polarization

$$P = \chi^{(3)} E_1 E_S^* E_2$$

arises and a conventional version of the phase conjugation effect takes place under condition for the four-wave mixing which corresponds to the first-order Bragg diffraction from the recorded dynamic grating. The diffracted wave E_D travels exactly against the signal wave E_S irrespective of the signal wave propagation direction. However, the phase-matching condition (the condition for Bragg diffraction) can be also met in the second (or higher) diffraction order by changing the propagation direction of the reading wave E_2 [12]. In this case, the diffracted wave E_D is determined by the nonlinear polarization

$$P = \chi^{(N-1)}(E_1 E_S^*)^M E_2,$$

(M is the diffraction order), and we deal with the N-wave coupling ($N = 2(M + 1)$) due to the $N - 1$-order nonlinearity. The reading waves E_2 should be directed into the nonlinear medium not exactly in the opposite direction, but at the angle

$$\gamma_2 = \arcsin(M\sin\gamma_1) - \gamma_1$$

with respect to the direction of propagation of the reference wave E_1 ($\gamma_2 \approx (M - 1)\gamma_1$for small angles γ_1). The propagation direction of the diffracted wave E_D is determined, in this case, from the phase-matching condition for the wave vectors

$$\mathbf{k}_D = M\mathbf{k}_1 - M\mathbf{k}_S + \mathbf{k}_2.$$

We will analyze the multiwave mixing using as an example recording of transmission dynamic gratings in resonant media, modeled by a two-level system. The system of equations that describes the formation of the wave E_D upon diffraction of the wave E_2 from the grating formed by the waves E_1 and E_S in a steady-state regime of the N-wave coupling ($N = 4, 6, 8, \ldots$) can be written in the form [12]

$$\left(\frac{\partial}{\partial z} \mp \gamma_1 \frac{\partial}{\partial x} + \frac{\Delta_\perp}{2ik}\right) E_{1,S} = \frac{i2\pi\omega}{cn_0}(\chi_0 E_{1,S} + \chi_{\pm 1} E_{S,1}), \quad (1)$$

$$\left(\frac{\partial}{\partial z} \pm \gamma_2 \frac{\partial}{\partial x} + \frac{\Delta_\perp}{2ik}\right) E_{2,D} = -\frac{i2\pi\omega}{cn_0}(\chi_0 E_{2,D} + \chi_{\mp M} E_{D,2}). \quad (2)$$

In these equations, γ_1 and γ_2 are the angles between the z axis and the wave vectors $\mathbf{k}_1$, $\mathbf{k}_S$ and $\mathbf{k}_2$, $\mathbf{k}_D$, respectively; $M = (N - 2)/2$ is the diffraction order; $k = \omega n_0/c$ is the wave number; n_0 is the nonresonant component of the refractive index of the medium; and $\Delta_\perp = \partial^2/\partial x^2 + \partial^2/\partial y^2$ is the transverse Laplacian.

The Fourier components of the nonlinear susceptibility of the medium

$$\chi_n = \frac{1}{2\pi}\int_{-\pi}^{\pi} \chi(\omega)\exp[-in((\mathbf{k}_1 - \mathbf{k}_S)\mathbf{r})]d((\mathbf{k}_1 - \mathbf{k}_S)\mathbf{r}),$$

$$n = 0, \pm 1, \pm 2, \ldots$$

are determined by the optical and spectroscopic characteristics of the resonance transition, as well as by the frequency and intensity of the interacting waves. When the two-level model of the resonant medium is used, the nonlinear susceptibility can be represented in the form [13]

$$\chi(\omega) = (n_0\kappa_0/2\pi)[\hat{\Theta}_{12}/B_{12} - \hat{\alpha}I/(1 + \alpha I)].$$

The complex parameter of the nonlinearity is given by the expression

$$\hat{\alpha} = a + i\alpha = (\hat{\Theta}_{12} + \hat{\Theta}_{21})/v p_{21},$$

where

$$\hat{\Theta}_{kl}(\omega) = \Theta_{kl}(\omega) + iB_{kl}(\omega).$$

The coefficients $\Theta_{kl}(\omega)$ are connected by the dispersion relations with the Einstein coefficients $B_{kl}(\omega)$ for induced transitions between the levels k and l, p_{21} is the total probability of spontaneous and nonradiative transitions, $v = c/n_0$ is the speed of light in the medium, and κ_0 is the linear extinction coefficient. The intensity of the light waves is normalized to the saturation intensity of resonance transition

$$I_{\text{sat}} = \alpha^{-1}, \quad \alpha = (B_{12} + B_{21})/v p_{21}.$$

In what follows, we will consider the case of coincident profiles of the absorption and luminescence ($\hat{\Theta}_{12} = \hat{\Theta}_{21}$).

In the approximation of a weak reading wave E_2 with respect to the waves E_1 and E_S that record the dynamic grating, the expressions for spatial components of modulation of the nonlinear susceptibility have the following form [11]:

$$\chi_0(\omega) = \frac{n_0\kappa_0}{2\pi}\left(\frac{\hat{\Theta}_{12}}{B_{12}} + \frac{\hat{\alpha}}{\alpha}\frac{1 - A_0}{A_0}\right), \quad (3)$$

$$\chi_{\pm 1}(\omega) = \frac{n_0\kappa_0}{2\pi}\left\{-\frac{2\hat{\alpha}\sqrt{I_1 I_S}}{A_0[1 + \alpha(I_1 + I_S) + A_0]}\right\} \times \exp[\pm i(\varphi_1 - \varphi_S)], \quad (4)$$

$$\chi_{\pm 1}(\omega) = \frac{n_0\kappa_0}{2\pi}\frac{\hat{\alpha}(-2\alpha\sqrt{I_1 I_S})^M}{\alpha A_0[1 + \alpha(I_1 + I_S) + A_0]^M} \times \exp[\pm iM(\varphi_1 - \varphi_S)], \quad (5)$$

where

$$A_0 = [1 + 2\alpha(I_1 + I_S) + \alpha^2(I_1 - I_S)]^{1/2}.$$

The studies of the transformation of the spatial structure of a light beam under conditions of multiwave mixing in resonant media imply numerical solution of the system of wave equations (1) and (2) with an allowance for the explicit form of the expressions for the Fourier components of the nonlinear susceptibility (3)–(5).

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 the 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 profiles of the signal and reference waves in the bulk of the nonlinear medium [direct solution of Eq. (1)], then we solved the total system of equations (1) and (2) backward from the boundary $z = L$ to $z = 0$ and found spatial profiles of the reading and diffracted beams.

In the numerical simulation, we assumed that the signal light beam E_S contained a screw phase dislocation of the topological charge m:

$$E_S(z = 0, r, \varphi)$$
$$= E_{S0}[(r - r_S)/r_0]^{|m|} \exp[-(r - r_S)^2/r_0^2 + im\varphi].$$

As the reference (E_1) and reading (E_2) beams, we used those with a plane wavefront and Gaussian amplitude profile: $E_1(z = 0, r, \varphi) = E_{10}\exp[-(r - r_1)^2/r_{01}^2]$ and $E_2(z = L, r, \varphi) = E_{20}\exp[-(r - r_2)^2/r_{02}^2]$. To provide efficient overlap of the beams in the bulk of the medium, the half-widths of the reference and reading beams were selected as four times larger than that of the signal one ($r_{01} = r_{02} = 4r_0$). The light beams intersected in the nonlinear layer at the angle $2\gamma_1 = 40$ mrad, the initial distance between their centers at the boundary $z = 0$ being $r_1 - r_S = r_0$. The half-width of the signal light beam was taken to be $r_0 = 1$ mm, the peak intensity of the reference beam was varied within the range $\alpha I_0 = 1$–5, the light wavelength ($\lambda = 0.5$ μm) coincided with or was close to the center of resonance absorption band of the nonlinear medium, the initial absorption coefficient was $k_0 = 1$ cm^{-1}, and the thickness of the nonlinear layer was varied within the range 1–3 cm.

Consider first the case when the frequency ω of the interacting light waves coincides with that of the resonance transition in the nonlinear medium. Under these conditions, interference of the light beams in the nonlinear layer results in formation of the amplitude holographic grating of absorption coefficient with no modulation of the refractive index.

An analysis of Eq. (2) and expression (4) shows that, in the process of four-wave coupling with the use of the plane reference and reading waves ($\varphi_1 + \varphi_2 = \text{const}$), the phase of the wave diffracted into the first order is opposite to that of the signal wave ($\varphi_D = -\varphi_S$, the phase conjugation effect). When the signal beam contains an m-order screw phase dislocation, the wavefront of the conjugate light beam must contain dislocation of the opposite sign ($-m$). At the same time, as follows from expression (5), diffraction into the second and higher orders in the appropriate configuration of the multiwave mixing allows one to implement operations of multiplication of wavefront phase inhomogeneities of the signal light beam E_S. For the plane reference and reading waves ($M\varphi_1 + \varphi_2 = \text{const}$), the phase of the diffracted wave becomes multiple of that of the signal wave with opposite sign ($\varphi_D = -M\varphi_S$). This makes it possible to invert the sign of the topological charge of

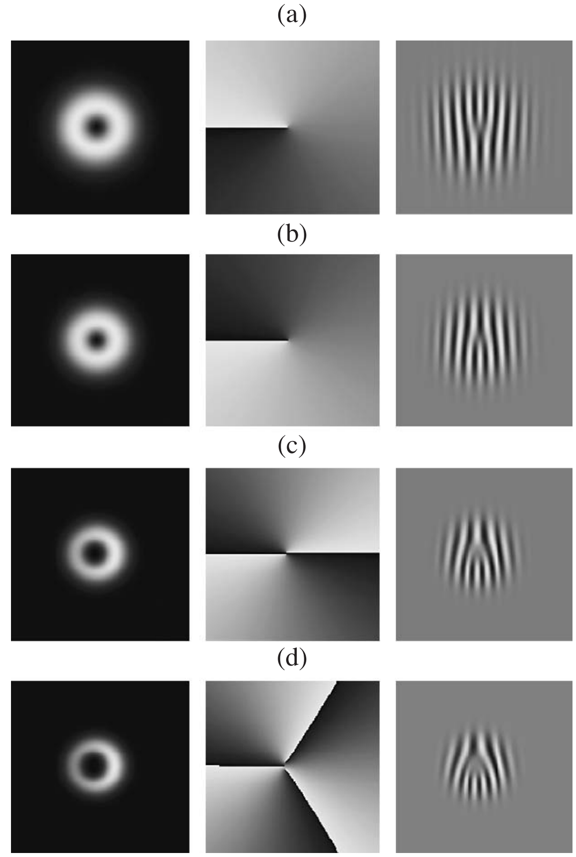

Fig. 1. Distribution of the intensity and phase in the transverse section of the (a) signal and diffracted light beams for the (b) four-, (c) six-, and (d) eight- wave interactions at the boundary $z = 0$ and the interferograms characterizing their topological structure; $\eta = 0$, $k_0L = 1$, $\alpha I_1 = 1$, and $\alpha I_S = 0.1$.

the singular light beam and simultaneously to multiply it by the parameter M that specifies the order of the Bragg diffraction.

The results of numerical simulation of the system of Eqs. (1) and (2) under the four-wave mixing condition are shown in Figs. 1a and 1b. As one can see, due to diffraction of the reading wave E_2 from the amplitude grating written by the reference beam E_1 and signal beam E_S, containing screw dislocation of the topological charge $m = 1$, a conjugate wave E_D is formed whose wavefront contains the dislocation of opposite sign ($m = -1$). This fact is also confirmed by the structure of the interferogram of the signal and diffracted beams with the plane reference wave (right column in Fig. 1).

In the numerical simulation of the multiwave mixing configurations, we used the same parameters of the nonlinear medium and radiation that were used for the case of four-wave mixing, except for the angle between the reading and diffracted light beams $2\gamma_2$. In conformity with the phase-matching conditions, $2\gamma_2 = 80$ and 120 mrad for the six- and eight-wave interaction, respectively. The results of a numerical analysis of the

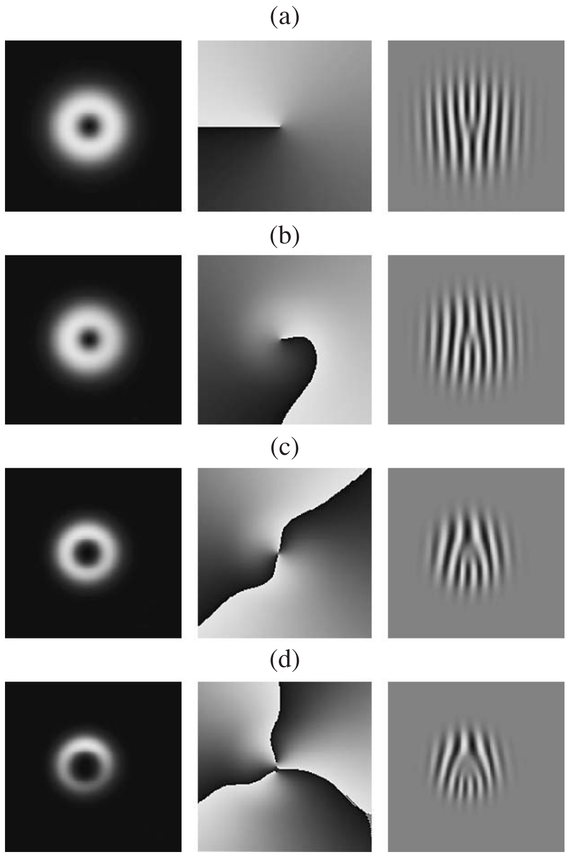

Fig. 2. Distribution of the intensity and phase in the transverse section of the (a) signal and diffracted light beams for the (b) four-, (c) six-, and (d) eight- wave interactions at the boundary $z = 0$ and the interferograms characterizing their topological structure; $\eta = -1.5$, $k_0L = 1$, $\alpha I_1 = 1$, and $\alpha I_S = 0.1$.

system of Eqs. (1) and (2) under conditions of six- and eight-wave interactions are shown in Figs. 1c and 1d. It follows from this that implementing six- and eight-wave interactions makes it possible to invert the sign of the topological charge of the optical vortex ($m = 1$) with simultaneous doubling ($m = -2$, Fig. 1c) or tripling ($m = -3$, Fig. 1d) of the topological charge.

Detuning the light wave frequency ω with respect to the absorption band center of the resonant medium gives rise to the self- and cross-modulation effects upon propagation of the light beams in the bulk of the medium. In this case, the efficient recording of the dynamic phase gratings is possible due to spatial modulation of the nonlinear medium refractive index [14]. However, the wavefront of the singular light beams is being distorted due to the effects of self-focusing or defocusing [15], which may deteriorate quality of the phase-matching.

The simulation of the four- and multiwave mixing processes for the singular light beams was performed with the light wave frequency ω shifted toward lower frequencies from the absorption band center of the medium ($\eta \equiv (\omega - \omega_{12})/\Delta = -1.5$, where Δ is the half-width of the Gaussian profile of the absorption band. For the chosen parameters, the refractive index of the resonant medium is efficiently modulated, while the conditions for the modulation instability are absent. The results of numerical calculations of spatial distribution of the intensity and wavefront structure of the light beams, and the corresponding patterns of interference with a plane wave are shown in Fig. 2. The diffracted light beams are seen to be characterized by the wavefront distorted with respect to the signal beam. A phase shift arises whose value is determined by the ratio of contributions from modulation of the absorption coefficient and refractive index to the nonlinear susceptibility and, for the case of the four-wave mixing, it can be evaluated using the formula

$$\Delta\varphi = \arctan(a/\alpha) = \arctan(\theta_{kl}/B_{kl}).$$

In the above case, the value of the phase shift is $\Delta\varphi \approx \pi/2$ because of the detuned operating frequency and predominant contribution of the phase gratings to the efficiency of the multiwave mixing. In addition, the nonlinear variation of the refractive index results in an incomplete restoration of the wavefront structure because of the difference in the phase modulation signs for singular beams with different topological charges ($m = \pm 1$). The above distortions of the wavefront can be revealed in the interference patterns of diffracted beams.

For the higher-order optical vortices ($m \geq 2$), one must also analyze the problem of their stability upon propagation in a nonlinear medium [16]. As shown by the results of the performed numerical simulations, the main factor that determines the possibility of generating singular beams with topological charges $m = 2, 3, \ldots$ and so on, upon the multiwave mixing, is related to the efficient overlap of all the light beams in the bulk of the resonant medium. Since the reading and diffracted beams, in the general case, are not collinear with respect to the reference and signal beams, the decrease in the degree of overlap of the light beams in the nonlinear layer not only deteriorates the quality of the transformation of the singular signal beam, but also destroys its structure by forming several optical vortices with smaller topological charges. This conclusion is demonstrated by Fig. 3, which specifies the limits of stability of topological stricture of the diffracted beam with the topological charges $m = -2$ (Fig. 3a) and $m = -3$ (Fig. 3b) for the case of six- and eight-wave coupling, respectively. The calculations were performed for different values of the angles of convergence of the light beams γ_1, different lengths of the nonlinear layer L, and different detunings of the light wave frequency from the absorption band center of the resonant medium (curves *1–5*). All the rest parameters corresponded to Figs. 1 and 2. The regions of stability of the beam with the topological charge $m = -2$ for the six-wave coupling lie below the curves *1–5* in Fig. 3a. Note

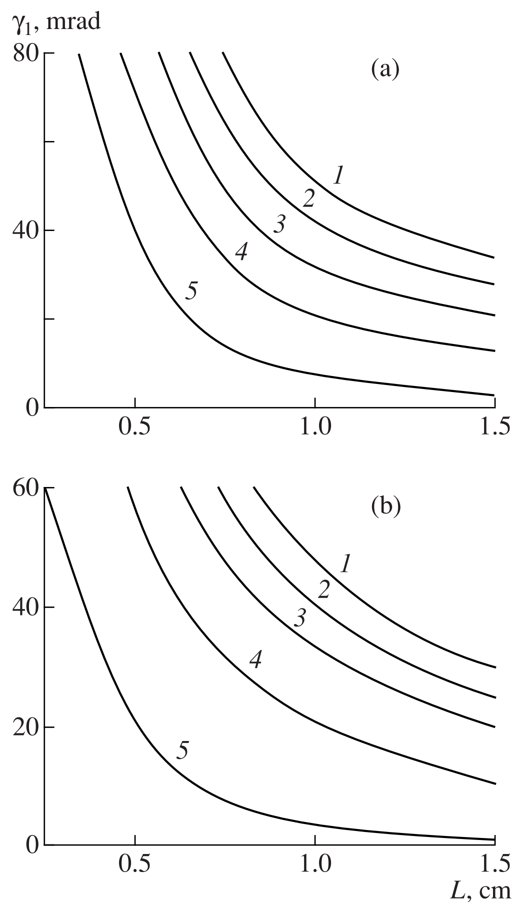

Fig. 3. Region of stability of the (a) second- and (b) third-order optical vortices created under conditions for the (a) six- and (b) eight- wave interactions; η = (*1*) 0, (*2*) 0.5, (*3*) 1, (*4*) 1.5, and (*5*) 2.

that an increase of the nonlinear modulation of the refractive index of the resonant medium with detuning of the frequency of the interacting waves from the absorption band center considerably narrows the range of stability for the higher-order optical vortices. As follows from Fig. 3b, to obtain optical vortices with $m = -3$ under conditions of the eight-wave coupling, one must satisfy even stricter requirements.

Thus, as the above theoretical analysis and numerical simulation show, the configurations of the multiwave mixing in resonant media can be used to change the sign of the topological charge of singular light beams with simultaneous multiplication by the parameter corresponding to the order of diffraction of the reading wave from the nonlinear dynamic grating. Analysis of stability conditions for the optical vortices arising in the six- and right-wave interactions shows that they can be implemented under typical experimental conditions.

In conclusion, the methods for multiplying the topological charge of singular light beams demonstrated in this paper by no means exhaust all possible mathematical operations with the use of topological charge of optical vortices characteristic of the multiwave mixing in resonant media. In particular, the introduction of phase singularity into the reference or reading beam noticeably complicates the pattern of interaction, but widens the possibility of control of the topological charge. An analysis of all possible combinations of interaction of the Gaussian and singular light beams is outside the scope of this paper.

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