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Energy exchange between coherent light beams with spatial phase distortions in an azopolymer film

M.S. Andreeva, V.I. Shmal'gauzen

***Abstract*. The energy exchange between coherent light beams with wave-front distortions in a photosensitive azopolymer film is experimentally studied. By using an adaptive interferometer with an azopolymer film as an element for combining the wave fronts of light beams, the phase visualisation is performed by transforming the spatial phase distortions of one of the beams incident on a medium to the amplitude distortions of another beam at the output. The experimental results agree with the theoretical conclusion that the spatial phase modulation at the input to a polymer is transformed to the amplitude modulation at the output and vice versa.**

Keywords: *azopolymer, two-wave interaction, adaptive interferometer, phase visualisation.*

1. Introduction

The study of photosensitive azopolymers attracts great attention due to their high photosensitivity, high radiation resistance, and the possibility to control their dynamic parameters with the help of external actions [1 – 3]. A change in the refractive index of such polymers is caused by reversible *trans – cis* isomerisation of azo dye molecules. Azo dye molecules irradiated by light undergo transitions from the stable *trans* state to the *cis* state. The reverse process can be induced by light or can occur spontaneously due to thermal relaxation. It was shown in [4] that azopolymers with liquid-crystal properties can be used as elements for combining wave fronts (light beam mixers) in adaptive interferometers. Adaptive interferometers of this type [5] differ from conventional interferometers, in which the wave fronts of light beams are superimposed on the output semitransparent mirror, in that they use a dynamic hologram written in a nonlinear material to mix light beams.

In this paper, we studied experimentally energy exchange between signal and reference light beams in an adaptive interferometer with a liquid-crystal polymer film and explained the observed effects based on the theory developed in [6]. We demonstrated experimentally that small spatial distortions of one of the beams interacting in a photosensitive azopolymer were transformed to the amplitude distortions of another beam.

M.S. Andreeva, V.I. Shmal'gauzen International Laser Center, M.V. Lomonosov Moscow State University, Vorob'evy gory, 119992 Moscow, Russia; e-mail: andreeva_maria@mail.ru, shm@lado3.ilc.msu.su

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2. Energy exchange between coherent light beams in azopolymer

We considered in [6] the interaction of light beams incident on a polymer film at different angles to the normal. The solution of this problem can be used to study energy exchange in the case of a signal beam with a distorted wave front. For this purpose, the beam with small wave-front distortions is represented as a sum of spatial spectral components with plane wave fronts inclined at different angles to the film surface. It is assumed that the distorted beam intensity is much smaller than that of the plane reference beam, so that the nonlinear interaction between spatial harmonics can be neglected. We studied a weakly distorted wave front of one of the beams, assuming therefore that the difference between the angles of incidence of the beams is much smaller than each of the angles, $\delta\theta \ll \theta$.

The geometry of interaction between the reference beam (R) and a spectral component (S) of the signal beam inclined at angles θ and $\theta + \delta\theta$ to the normal to the polymer plane is shown in Fig. 1. Polarisations of the interfering beams are assumed identical and perpendicular to the plane of incidence xz (the beams are s-polarised). The effect of light

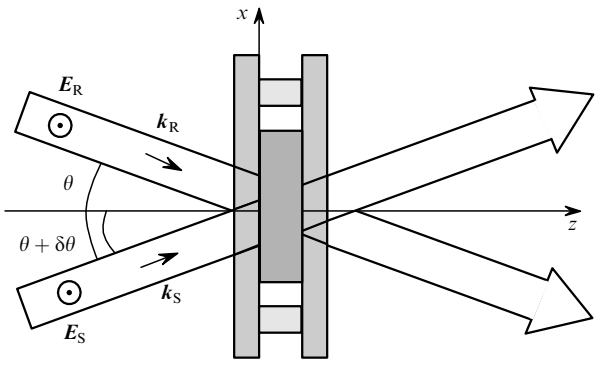

Figure 1. Scheme of the two-wave mixing of coherent light beams in a photosensitive azopolymer film. $\boldsymbol{E}_{R,S}$ and $\boldsymbol{k}_{R,S}$ are the electric field strengths and the wave vectors of the reference and signal waves interfering in the polymer film.

beams reflected from the substrate–air interface on energy exchange is neglected.

Because a photosensitive polymer has a local response to the action of light, energy exchange can occur in it either in the transient regime (at the relaxation time scale) or in the stationary regime if the frequencies of interacting waves are different [4].

According to the assumption that the signal beam is much weaker than the reference beam, the theory developed in [6] used the interference pattern small-contrast approximation. In this approximation and for small inclination angles of one of the beams, the intensity and refractive-index gratings can be considered harmonic. In the problem of energy exchange between the beams having the same inclination angles with respect to the normal to the polymer plane, the vector of the intensity grating is oriented in the plane of a polymer layer. Because the beams have different inclination angles, the spatial period of the intensity grating changes, resulting in a change in the modulation depth of the refractive-index grating due to a finite spatial resolution of the medium. A change in the inclination of one of the beams in the plane of incidence affects the period of the refractive-index grating much stronger than the inclination in the perpendicular plane, which only slightly changes the period. Therefore, we will take into account the angular mismatch only in the plane of incidence of light beams. Because the signal and reference beams diffract from the same refractive-index grating written by them in the polymer, the amplitude distortions at the film output will be revealed in both beams.

The energy-exchange equations for the case of symmetrically incident beams in [4] are valid for different photosensitive media, being different only in the form of the function $\Delta n^{(1)}$ (the modulation depth of the refractive-index grating). The form of the function $\Delta n^{(1)}$ for a particular medium can be found both theoretically, by using different models, and experimentally. In the case of beams incident at different angles, one should take into account that the modulation depth of the refractive-index grating depends both on the intensities of the signal and reference beams and the difference of angles of incidence. In the steady-state regime, the energy-exchange equations for the frequency-shifted (by Ω) beams take the form

$$\frac{\partial I_S}{\partial z} = -2I_S \mathrm{Im}[\Delta n^{(1)}(I_S, I_R, \delta\theta)],$$

$$\frac{\partial I_R}{\partial z} = -\frac{I_R(2I_0 - I_R)}{I_0}\mathrm{Im}[\Delta n^{(1)}(I_S, I_R, \delta\theta)], \quad (1)$$

$$\frac{\partial \xi}{\partial z} = B_1 \tan\theta\,\delta\theta + \mathrm{Re}[\Delta n^{(1)}(I_S, I_R, \delta\theta)].$$

Here, I_0 is the incident beam intensity; $I_{R,S}$ are the intensities of the reference and signal beams; the coefficient B_1 is proportional to the stationary value of the photoinduced addition to the refractive index of the polymer; ξ is the phase difference between the signal and reference beams. One can see from (1) that a small angular mismatch $\delta\theta$ explicitly affects only the phase shift between the beams. Taking into account spatial diffusion, describing a finite spatial resolution of the polymer film, $\Delta n^{(1)}$ also depends on the difference of angles of incidence, but this dependence is weak.

To study the behaviour of the amplitude and phase of different spectral components in the case of a constant frequency detuning of the interacting beams, it is convenient to consider the phase shift $\Delta\xi = \xi(\delta\theta) - \xi(\delta\theta = 0)$ and the normalised intensity $I_S^{\mathrm{norm}} = I_S(\delta\theta)/I_S(\delta\theta = 0)$.

The expressions for these quantities at the output of a thin layer of thickness z_0 can be readily obtained from system (1) by expanding in a series in small angle $\delta\theta$:

$$I_S^{\mathrm{norm}} = \exp[c(\Omega)\delta\theta z_0], \quad (2)$$

$$\Delta\xi = [2d_1(\Omega) + d_2(\Omega)\delta\theta]z_0, \quad (3)$$

where the coefficients $c(\Omega)$, $d_1(\Omega)$, and $d_2(\Omega)$ can be calculated by using equations from [6], which are not presented here due to the cumbersome expressions. For small angles $\delta\theta$, the amplitude and phase of each spectral component linearly depend on the mismatch angle (only the first term in the expansion in powers of $\delta\theta$ is taken into account).

Thus, the frequency shift between the signal and reference beams, taking into account diffusion terms in the equation for the photoinduced refractive index, results in the dependence of the intensity and phase of the signal beam at the output of the polymer film on the difference of the angles of inclination of the interacting beams and the frequency difference of the beams. A similar dependence is also revealed in the transient process upon switching on or off one of the beams. Such an effect in the case of the signal beam with a distorted wave front leads to the transformation of phase inhomogeneities in the signal beam to the amplitude distortions in the reference beam and can be used for studying the interaction between the beams with nonplanar wave fronts. To analyse these processes qualitatively, it is sufficient to take into account a linear dependence of the phase and amplitude logarithm on the mismatch angle. The effective transformation length of phase distortions to amplitude distortions depends on the characteristic scale of phase distortions (in experiments, inhomogeneities of size 0.5 mm were used).

3. Experimental observation of the transformation of phase distortions to amplitude distortions

To verify the theoretical results obtained in [6], we studied experimentally the transformation of spatial wave-front phase distortions of one of the beams interacting in the azopolymer film to the amplitude distortions of another beam during the transient process. The scheme of the experimental setup (adaptive interferometer) is presented in Fig. 2. The radiation wavelength was selected according the spectral properties of the polymer film. The SPK-4/4 azopolymer (20 % of the dye) studied in the paper had the absorption maximum in the 350–400-nm region [7]. The maximum change in the refractive index of the polymer is achieved upon irradiating it by light in a longer-wavelength region. Holograms are produced and stored by using, as a rule, different light sources. To control the properties of the medium, the wavelength of light should lie within the absorption band of the polymer. In the problems of adaptive interferometry, one radiation source is used; therefore, to provide the efficient writing of dynamic holograms, it should have the wavelength that lies within

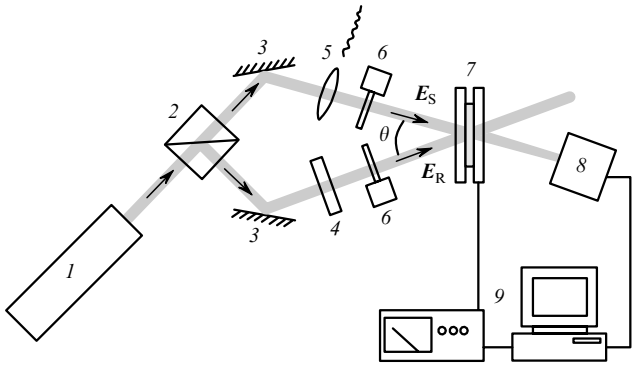

Figure 2. Scheme of the experimental setup: (*1*) frequency-doubled Nd : YAG laser; (*2*) beamsplitter cube (1 : 10); (*3*) mirrors; (*4*) compensator; (*5*) object introducing spatial phase distortions (lens or etched glass plate); (*6*) shutters; (*7*) cell with a polymer; (*8*) CCD camera; (*9*) experiment control system.

the absorption band of the polymer but at the same time is not strongly absorbed by the sample.

We used in experiments the second harmonic of Nd : YAG laser (*1*), which was incident on beamsplitter cube (*2*) with the splitting ratio 1/10. The power density of the reference beam incident on a medium was 60 mW cm^{-2}, the power of the signal beam was an order of magnitude lower. The light beam reflected from mirrors (*3*) intersected at an angle of $\theta \simeq 15^\circ$ in polymer sample (*7*) by producing an interference pattern. The thickness of a polymer film was 50 μm, which required its optimisation due to strong absorption of light in the polymer. The thickness of glass substrates (4 mm) between which the polymer was placed was much greater than that of the film. Control system (*9*) contained a thermal stabilisation unit and a generator. Spatial phase distortions were introduced by object (*5*) placed into the signal beam. The regular distortion of the wave front of the signal beam was introduced by a positive eyepiece 1.5-diopter lens, while irregular phase distortions were produced with an etched glass plate with random phase inhomogeneities of size ~ 0.5 mm. The beams had the same linear s-polarisation and could be 'switched off' by shutter (*6*). The temperature of the polymer film was controlled with a thermal stabilisation unit. The orienting alternating electric field with the frequency 350 Hz and the amplitude ~ 50 V was applied to the sample with the help of a system of transparent electrodes. The intensity distribution in the signal beam at the output of the polymer film was recorded with CCD camera (*8*). The output video signal of the CCD camera was fed to the input–output video board of a computer and then was processed by using a special software package.

Figure 3a shows the intensity profile of the signal beam transmitted through the polymer film when a lens was place in the signal beam. One can see that the beam is focused compared to the beam recorded without the lens (Fig. 3b). The signal beam was blocked at some instant; however, because of the photoinduced mismatch between the refractive-index and intensity gratings, the diffraction of the reference beam was observed in the signal-beam direction during the relaxation time, the decay time being dependent on the external conditions (temperature, the intensity ratio of the beams, and the total radiation power density). Figures 3c–f show the intensity distributions over the cross section of the reference beam diffracted in the signal-beam direction. One can see that the intensity distribution of the diffracted reference beam almost coincides with that of the signal beam with the distorted wave front before its switching off. Therefore, this experiment shows how the phase distortions of the signal beam are transformed to the amplitude distortions of the reference beam due to energy exchange.

Figure 4 presents the intensity profile of the signal beam in which an etched glass phase plate with irregular phase distortions was introduced. As in the experiment with regular distortions, the signal beam was switched off at

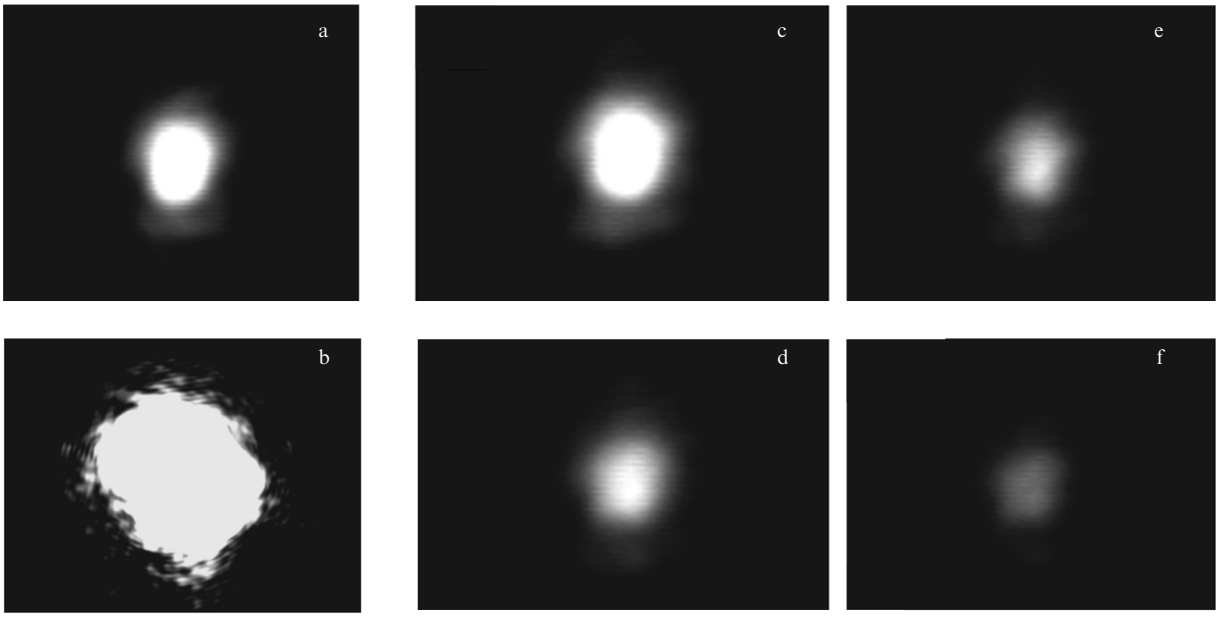

Figure 3. Intensity distributions of the signal beam after (a) and before (b) focusing, and the intensity distributions of unfocused reference beam diffracted in the signal-beam direction at different instants (c–f).

some instant and the diffraction of the reference beam in the direction of signal beam was observed. Figure 4b clearly shows that the type of amplitude distortions in the beam diffracted in the direction of the signal beam is the same as in the signal beam (Fig. 4a), which demonstrates the visualisation of phase distortions. To confirm the regularity of these effects, we performed experiments for different regions of the phase plate. Figures 4c, d show the results of similar experiments for the phase-plate region containing other phase inhomogeneities. One can see that the visualisation effect is observed for different phase inhomogeneities.

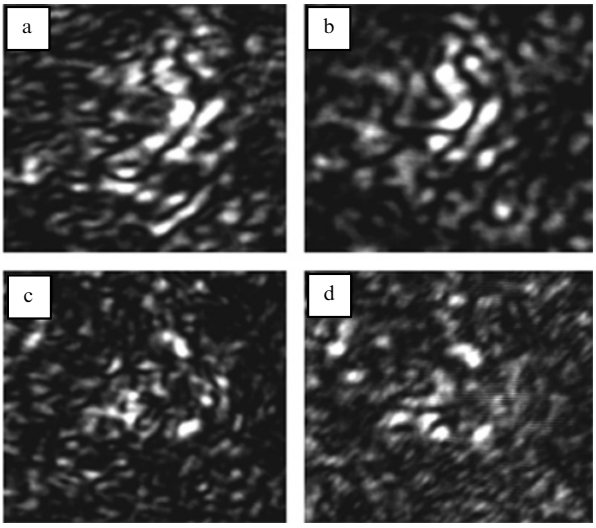

Figure 4. Intensity distributions of the signal beam with random wave-front distortions for different regions of a phase plate (a, c), and the intensity distributions of the beam diffracted in the signal-beam direction (b, d) for regions (a) and (c), respectively, when the signal beam is blocked.

4. Conclusions

We have shown that the intensities of spectral components incident at different angles to the normal in an adaptive interferometer with a polymer film are differently transformed after transmission through the film. For this reason and due to energy exchange between the beams in the film, the wave-front phase distortions of the signal beam are transformed to the amplitude distortions of the reference beam (the phase visualisation is observed). This effect can be explained as follows: if the wave front of one of the beams is distorted, the refractive-index grating in the polymer is also distorted. The reference beam with a plane wave front diffracts from the distorted grating and acquires in turn amplitude distortions, carrying information on phase distortions in the signal beam.

Therefore, we have shown experimentally that in an adaptive interferometer with a photosensitive azopolymer film as a mixer of light beams, the spatial phase modulation of the signal beam is transformed to the amplitude modulation of the reference beam.

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