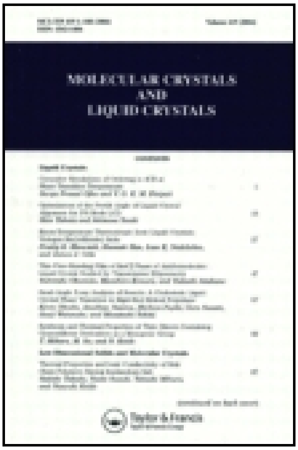

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Vector Holograms Recorded in Epoxy-Based Polymer With 4-Aminoazobenzene by Pulsed Laser Light

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Both scalar and vector gratings were recorded in an epoxy-based polymer with 4-amino-azobenzene depending on the geometry of the recording by the second harmonic of a pulsed Nd:YAG laser. A new mechanism of nonlinear refraction (striction) is suggested and is experimentally observed for the first time in the medium under study.

Keywords: holographic gratings; nonlinear refraction; striction gratings

1. INTRODUCTION

The polarization or vector holograms makes it possible to record and to restore not only the amplitude and the phase of interacting waves, but also their state of polarization. For recording the vector holograms, it is necessary to have a recording medium with anisotropic nonlinear optical properties. Vector holograms have been recorded for the first time in 1972 using the absorption dichroism of F_A-centers in alkali halide crystals (the Weigert effect) [1]. After that, photochromic glasses [2], solutions of organic dyes [3], bacteriorhodopsin [4], and

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semiconductors, namely CdS [5], were used as recording media for the polarization recording. It is also known that the vector hologram recording is a powerful technique for the study of mechanisms of nonlinear optical response of the matter.

Earlier, we have studied the nonlinear refraction in an epoxy based polymer modified by amino azobenzene under pulsed laser excitation [6]. We observed the strong third-order optical nonlinear response in this material excited by nanosecond pulses of the second harmonic of a Nd:YAG laser. The nonlinear refraction relaxation time covers a wide range from nanoseconds to days depending on the excitation intensity due to the contribution of many physical mechanisms of changes in the nonlinear refraction index. The analysis of possible nonlinear refraction mechanisms in the medium in question carried out on the basis of our studies showed that the anisotropic nonlinear mechanisms such as the orientation and isomerization of azobenzene molecules can take place. Our further studies by means of the polarization technique [7] showed that an anisotropy of the nonlinear refractive index is induced by the second harmonic of a YAG:Nd^{+3} laser in the medium under study. Such an anisotropy can be used for the vector hologram recording.

The aim of this work is to determine the possibility to record vector holograms in an epoxy-based polymer with covalently attached molecules of asobenzene and to investigate the anisotropic mechanisms of nonlinear refraction in the medium by means of the polarization holography.

2. SAMPLE

The molecular structure of our polymer is shown in Figure 1. An aminoazobenzene (AA) molecule is attached to the polymer chain (bisphenol-A diglicydyl ether) by a covalent bond.

Figure 1 shows also the absorption spectrum of the polymer in the visible region. The intense long-wavelength absorption band with a maximum at 430 nm of the azobenzene trans-isomer corresponds to the n-π^* transition [8,9]. As a result of these transitions, the trans-cis isomerization takes place. The arrow in Figure 1 indicates a wavelength of 532 nm, at which the sample was excited at a long-wavelength of this absorption band. The experimental samples were in the form of a thin polymer layer with a thickness of 100 μm placed between two glass plates. The glass transition temperature T_g of the sample was 100°C.

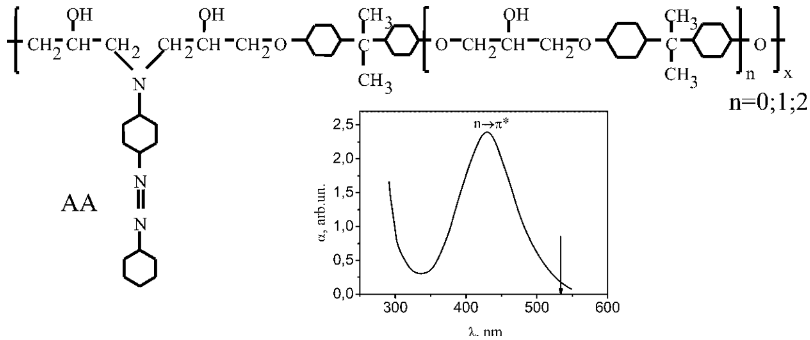

FIGURE 1 Molecular structure of the epoxy-based linear polymer with 4-aminoazobenzene and its absorption spectrum.

3. EXPERIMENTAL SETUP

In the intensity or scalar holography, the recording medium is exposed to an interference pattern formed by two coherent light beams with parallel polarization. In such a case, the light intensity modulation is produced at the intersection of the recording beams (Fig. 2a). In the polarization holographic recording, two waves have orthogonal polarizations so that the resultant light intensity is constant, but the light polarization has periodic space modulation in accordance with the phase difference between the two recording beams (Fig. 2b).

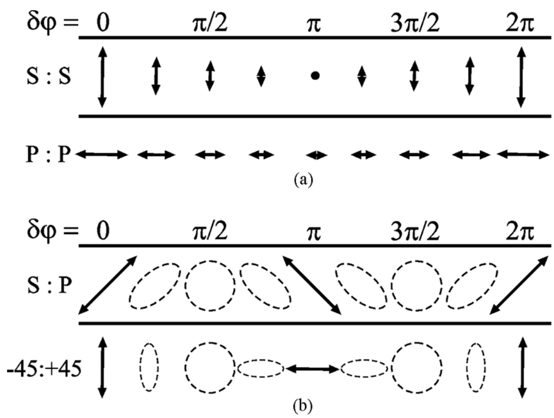

FIGURE 2 Space distribution of the resultant light polarization for different geometries in accordance with a phase difference between two recording beams.

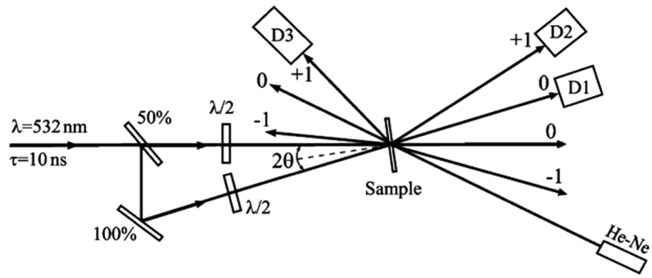

FIGURE 3 Experimental setup. D1, D2, and D3 are photodetectors; a frequency-doubled one-mode Nd^{3+}:YAG laser ($\lambda = 532\,\text{nm}$, $\tau_p = 10\,\text{ns}$, TEM_{00}); a cw He–Ne laser.

The scheme of the experimental setup is shown in Figure 3. As a source of irradiation, we used a single-mode frequency-doubled pulsed YAG:Nd^{+3} laser ($\tau_p = 10\,\text{ns}$, $\lambda = 0.532\,\mu\text{m}$, $E_p = 1.5\,\text{mJ}$, TEM_{00}). Two $\lambda/2$ plates were used to obtain different recording geometries such as S:S (when the electric field E of light waves is vertical), P:P (E is horizontal), S:P and -45°:$+45^\circ$ (when the vectors E of two recording waves are directed at 45° to the vertical position from different sides).

The diffraction efficiency of recorded holographic gratings was measured as the ratio of light intensities of the first and zero diffraction orders. The dynamics of the recording and relaxation of the gratings recorded by a single pulse of a YAG:Nd^{+3} laser was measured by reading out the gratings by a CW He–Ne laser radiation. That is, the time dependence of the He–Ne radiation intensity diffracted in the first order corresponds to a nonlinear optical response of the recording medium. The sample was shifted from record to record, so that every new grating was recorded in a new area of the sample to avoid the influence of a previous grating upon a new one.

4. EXPERIMENTAL RESULTS

Actually, the polarization gratings were recorded in the S:P geometry at $\lambda = 532\,\text{nm}$ in our samples [10]. We have shown also that the state of polarization of the recording beams was restored in the corresponding diffraction orders, which is a direct manifestation of the vector character of recorded gratings (see Fig. 4). The dynamics of the grating recording was qualitatively similar to that of the induced anisotropy. So we have come to a conclusion that the mechanisms of the optical nonlinear response in both cases are the same.

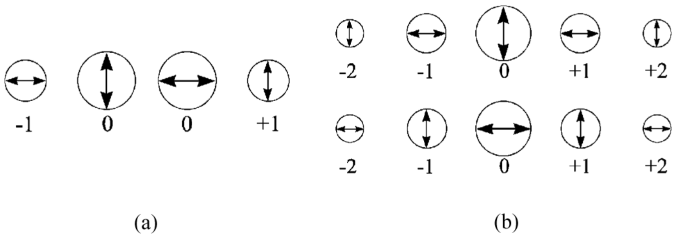

FIGURE 4 The state of polarization: a) of the recording and diffracted beams under self-diffraction; b) under the reading out of the grating by a measuring light source.

We have recorded the polarization gratings in different recording geometries mentioned above. However, the dynamics of nonlinear responses is different in spite of the type of recorded gratings: the scalar or vector one (Fig. 5). The dynamical curves for the scalar P:P and vector -45°:$+45^{\circ}$ geometries have similar shape: we observe the sharp leading edge of the diffraction efficiency and then the relaxation with a characteristic time of about 150 μs. The dynamical curves in two other geometries (the scalar S:S and vector S:P ones) also have similar shapes. The leading edge of the diffraction efficiency is slower than that in the previous case about 100 μs, and then the diffraction efficiency reaches its saturated value. Thus, the shape of the diffraction efficiency dynamics depends upon the recording geometry as a result of different mechanisms responsible for the recording.

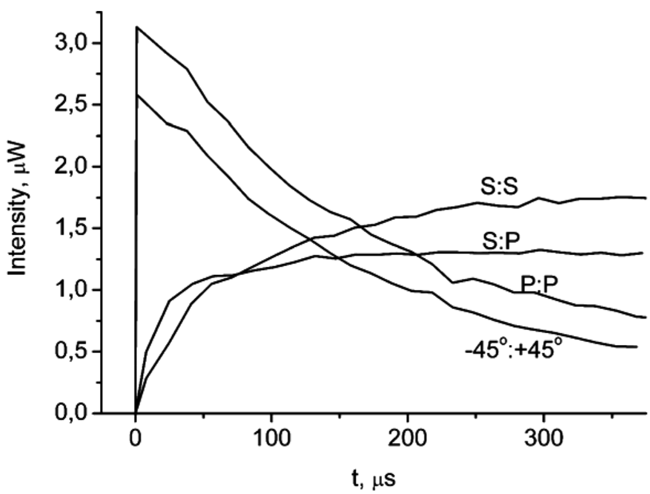

FIGURE 5 Dynamics of the scalar and vector gratings: build-up and relaxation.

As for the recording geometries P:P and -45°:$+45^{\circ}$, in our opinion, the striction or pressure gratings can be effectively recorded in our samples. It is connected with the particular features of our samples. They consist of a polymer layer between two glass plates, so the polymer has no free surface. However, when a polymer has free surface as in [11], the efficient relieve gratings were recorded just in the same geometries P:P and -45°:$+45^{\circ}$as a result of the polymer mass movement. In our case, such a movement cannot take place because of the glass plates.On the other hand, local ponderomotive forces can arise, by resulting in the recording of striction gratings.

5. DISCUSSION

We have described the mechanisms of nonlinear refraction for the recording geometries S:S and S:P, when striction gratings are not recorded, in our previous paper in detail [10]. We have suggested three main mechanisms for the refractive index to be nonlinear. These are the "angular hole burning" connected with the *trans-cis* isomerization of asobenzene molecules oriented in parallel to the light electric field vector; the reorientation of asobenzene molecules in the field of a linearly polarized light wave; and the reorientation of polymer chain fragments.

Let us now dwell upon the P:P and -45°:$+45^{\circ}$ geometries. The main difference of the geometries can be seen in Figure 2 presenting the interference patterns. One can see from the figure that the gradient of the electric field E has different orientations for different geometries. For the striction grating to be induced, the electric field gradient should be oriented along the grating vector or perpendicularly to grating fringes. Indeed, the horizontal component of the electric field is absent in the S:S geometry. In the P:P geometry, the component is changed from zero to $2E_0$. In the S:P geometry, the component is small, and, in the -45°:$+45^{\circ}$ geometry, the horizontal component of the electric field is changed from 0 to $\sqrt{2}E_0$.

The situation for the P:P geometry is presented in Figure 6 in more details. The figure shows the modulation of the electric field of a light wave in the interference pattern, as well as the electric field gradient $\mathrm{d}E(x)/\mathrm{d}x$ which is shifted by $\pi/2$. It is well known [11] that the pondoromotive or striction force proportional to the product $E(x)*\mathrm{d}E(x)/\mathrm{d}x$ looks as

$$f_x = \varepsilon_0 \left[\chi_{xx} E_x \frac{\partial}{\partial x} E_x + \chi_{yx} E_x \frac{\partial}{\partial x} E_x + \chi_{zx} E_x \frac{\partial}{\partial x} E_x \right]$$

The force acts upon a material in correspondence with the interference pattern which is seen in Figure 5. The arrows show the direction of the

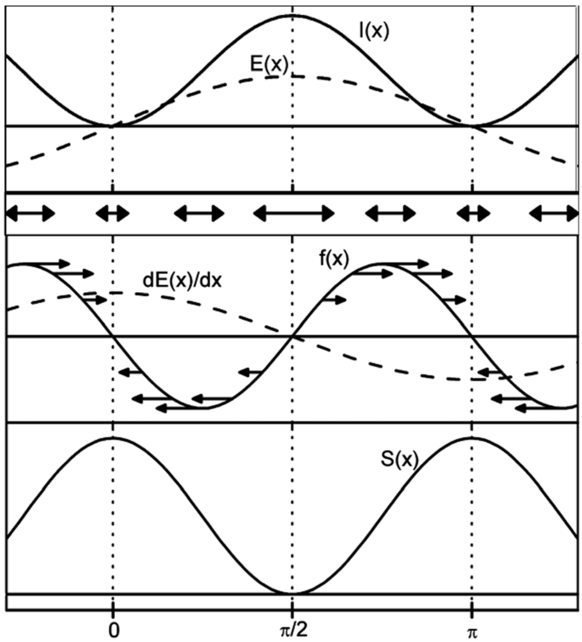

FIGURE 6 Space distribution of the intensity $I(x)$, electrical field $E(x)$, polarization $P(x)$, gradient of electrical field $dE(x)/dx$, and density $S(x)$.

force depending on the sign of $f(x)$. The material undergoes compression at the extremes of the gradient $dE(x)/dx$. The compression is periodic and forms the striction grating. The pressure grating is formed with the sound velocity. The grating period in our experiments was 6.5 μm, so the striction grating is induced during several nanoseconds, i.e., during a laser pulse (10 ns in our case). That is why the leading edge of the diffraction efficiency in the P:P and $-45°{:}+45°$ geometries is so sharp. It should be also underlined that the mechanical tensions suppress the reorientation of the fragments of polymer chains during the grating recording. In such a case, the relaxation of a recorded grating is faster, which is observed experimentally. Thus, it is possible to control the response time of a recording medium by using different recording geometries.

5. CONCLUSIONS

We have shown that both the scalar and vector gratings can be recorded by the second harmonic of a pulsed Nd:YAG laser in the epoxy-based polymer with 4-amino-azobenzene depending on the recording geometry. A new mechanism of nonlinear refraction is suggested and, for the first time, experimentally observed in the medium under study. It is shown that the striction gratings can be recorded in some recording geometries, when the light wave electric field gradient is parallel to the grating vector. We have also shown the possibility to control the recording medium response time just by changing the recording geometry.

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