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Photoinduced Optical Anisotropy in Azo-Dye Doped Polyimide Films*

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Using a two-wave mixing technique supplemented by the illumination of the second laser, we studied the dynamics of the photoinduced anisotropy in polyimide free-standing films doped by azo-dyes containing one or two N=N bonds. We have measured the characteristics of grating recording using a He-Ne laser source ($\lambda = 632.8$ nm) under influence of light coming from a cw YAG laser doubled in frequency ($\lambda = 532$ nm). The experiments in polyimide-Disperse Red 1 (4-[N-ethyl-N-(2-hydroxyethyl)]amino-4′-nitro-azobenzene) system revealed complex kinetics of photoinduced birefringence leading to grating recording and light self-diffraction. The two main processes were identified during the grating build-up in this high T_g polymer matrix. Preliminary results on possibility of dynamic color and polarization holographic recording in polyimide containing newly synthesized molecule having two N=N bonds 1,3 diamino-4-azo(4′-nitroazobenzene)benzene are also presented.

Key words: polyimide, holography, azo dye, molecular orientation, photoinduced reorientation

The guest-host system prepared from a small amount of highly anisotropic dye molecules dissolved in a polymer is an optically isotropic optical medium, providing the dye molecules are randomly distributed. If, in addition, the dye exhibits photochromic properties, *i.e.* can be switched by light of a suitable energy between its two stable forms, the system shows a photoinduced birefringence and dichroism. Exact molecular mechanism of such induced optical anisotropy depends on the dye optical properties as well as on the nature of the dye-matrix interactions. Classic systems are the polymeric matrices containing azo-dyes [1–7]. Most of azo-dyes, due to the nature of their N=N bond, exhibit photoisomerization of *trans-cis* and *cis-trans* type together with thermal relaxation process of *cis-trans* type as the *trans* state is usually more stable than the *cis* one. It has been shown and discussed in many papers [8–12] that a linearly polarized light incident on the thin layer of dye-polymer system induces an angular anisotropy to the initially isotropic distribution of the dye molecules through temporal or permanent molecular reorientations. Forced by light angular or-

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*Dedicated to the memory of Professor Krzysztof Pigoń.

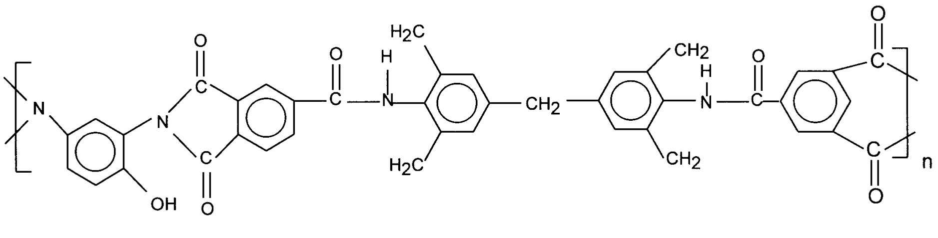
dering of the dye molecules (usually long molecular axes are aligned perpendicularly to the polarization plane of the light beam) and rotational diffusion are the two main competitive processes, which determine the effective photoinduced birefringence or dichroism of the system [13]. The photoinduced anisotropy, depending on conditions of light illumination (steady state or pulsed) and the nature of the matrix, can be quasi-permanent or transient – rapidly decaying when the light is switched off [14–18]. In most of the cases studied one observes the coexistence of the quasi-permanent and transient components of the photoinduced anisotropy, which suggests that the mechanism is relatively complex and dependent on ratios of time constants of involved molecular processes.

In the present work we study the properties of two systems composed of azo-dyes dissolved in a high glass transition temperature (T_g) polyimide. We will focus our attention to the rarely studied problem of the competitive molecular ordering induced by two different in frequency laser beams both falling within the absorption spectrum of the dye. The problem is related too with the photophysical hole burning or the optical information storage using wavelength multiplexing technique as well as potentially with the color real-time holography.

EXPERIMENTAL

Sample preparation: High temperature polycondensation method was used for the polymer preparation. A solution of equimolar amounts of diamidedianhydride (synthesized from trimellitic anhydride acid chloride and 4,4′-methylene bis(2,6-dimethylaniline) [19]) and 2,4-diaminophenol dichloride (1 mmol) in NMP and dichlorobenzene [80/20 (w/w), 20% total monomer concentration] was stirred at 175°C for 3.5 hours under argon atmosphere. Then the polymer solution was diluted with NMP and precipitated by slow dropping of the reaction mixture into methanol. The product was collected by filtration and further purified by Soxhlet extraction with methanol for about 24 hours. Then the polymer was dried at 150°C under vacuum for 24 hours. Synthesis of polyimide was carried out according to schemes described elsewhere [19,20]. Polymer loses 10% of weight in a nitrogen atmosphere at 410°C [19] and is soluble in a few solvents, like DMA, DMF, NMP and DMSO at room temperature. The molecular structure of the polyimide unit is shown in Scheme 1.

We have checked by X-ray diffraction studies that the polymer forms an amorphous matrix. Its glass transition temperature T_g is 322°C as detected by DSC. We have prepared two types of polymer foils doped with azo-dyes. One of them contains the well known 4-[N-ethyl-N-(2-hydroxyethyl)]amino-4′-nitro-azobenzene, Disperse Red 1 (abbreviated as DR1) and the second the newly synthesized 1,3 dia-



Scheme 1. The molecular structure of the polyimide unit.

mino-4-azo(4′-nitroazobenzene)benzene (abbreviated as AN) containing two diaza-groups. The chemical structures of both compounds are shown in Fig. 1. Foils have been obtained from the solution of the polymer (concentration of polymer was about 15% in DMF and amount of the dye ≅ 1 wt. %) by casting on glass plates and heated during 8 hours up to 200°C, then kept at 200°C in a vacuum overnight. Such a procedure yielded typical foils with thickness around 30 μm. Absorption spectra of polyimide foils containing 1% of DR1 and 1% of AN dyes are shown in Fig. 1. Both spectra were taken in reference to the pure polyimide foil.

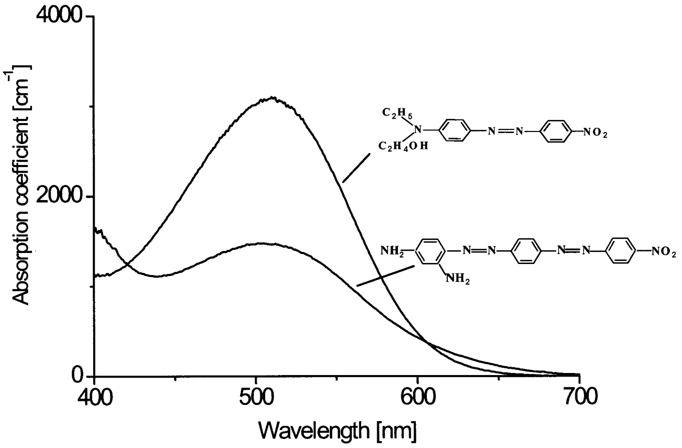


Figure 1. Absorption spectra of two azo-dyes dissolved in polyimide and measured with respect to the pure polyimide foil. Both foils have thickness 31 μm and contained 1 wt. % of the dye. The structural chemical formulae of DR1 (4-[N-ethyl-N-(2-hydroxyethyl)]amino-4′-nitro-azobenzene, the upper structure) and AN (1,3 diamino-4-azo(4′-nitroazobenzene)benzene, the lower structure) are shown.

Experimental setup: The setup for our experiment is presented in Fig. 2. A continuous wave (cw) doubled in frequency YAG laser (P = 25 mW) that provides linearly polarized single-mode emission (at 532 nm) was used for aligning the molecules of azo-dye in plane perpendicular to the polarization plane of the beam. A He-Ne laser (P = 25 mW) working at 632.8 nm was used for recording the holographic grating in the foils. The output beam from the laser was divided into two beams by cube beam splitter into equal power beams. Rotating the λ/2 plate placed on one of the beams allows for its polarization change, just we could obtain VV (vertical – vertical) or VH (vertical – horizontal) beams polarizations. For VV polarization the light intensity in the foil is periodically modulated:

$$I = I_0(1 + m\cos(Kx)) \tag{1}$$

where $I_0 = I_1 + I_2$, m is the modulation factor $m = (2\sqrt{I_1 I_2})/(I_1 + I_2)$ and $K = 2\pi/\Lambda$ is the grating wave-vector with Λ a grating period. The angle between two beams was 3.6° in air resulting in 7.2 μm grating spacing. For VH polarization the intensity of light incident on the sample is uniform $I_0 = const$ but there is a periodic modulation of polarization [13], which is sufficient for polarization grating recording in systems with photoinduced molecular reorientation [21].

Our measurements were performed as follows: We first recorded the holographic grating using He-Ne laser beams. As the grating strength (understood as the amplitude of light induced birefringence) in such dyed systems depends on the exposure time, the self-diffraction appears. We monitored the temporal behavior of the first order diffraction power in function of the beam polarizations using the power meter Labmaster Ultima (Coherent). By removing one of the recording beams we could study the relaxation of the grating. The recording and relaxation processes took several minutes before reaching saturation or complete grating removal. Then we studied the grating recording by He-Ne beams in the presence of the

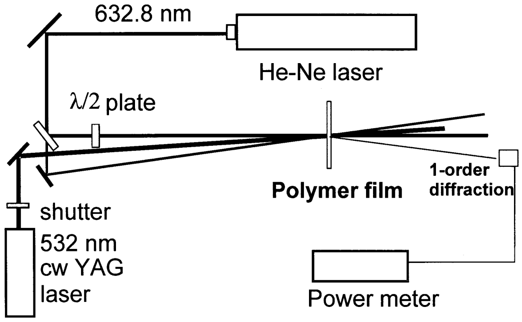


Figure 2. Experimental set-up for the measurements of the dynamics of grating recording in the polymer foils by the two-wave mixing technique in the presence of illumination by the second laser. A Spectra Physics He-Ne laser (P = 25 mW) emits light at 632.8 nm and a doubled in frequency DPSS 532 Coherent laser at 532 nm. The power diffracted into the first order diffraction direction is measured by the calibrated silicone detector and displayed by the Labmaster Ultima, Coherent power meter. The angle between hologram writing beams is 3.6°.

linearly polarized YAG laser illumination overlapping the region of the grating. Eventually we took off the YAG laser beam and studied the dynamics of grating reinforcement and its relaxation to the previous level when the YAG laser beam was switched on again.

RESULTS AND DISCUSSION

Description of the optical properties of the polyimide – azo-dye system: Let us consider a homogeneous thin film made of azo-dye-doped polyimide, that could be illuminated by light coming from two different laser sources *i.e.* with different wavelengths but both lying within the absorption band of the dye. In further discussion we will consider the well known molecule DR1, but general conclusion will be valid for those azo-dye molecules, which have two isomers: *cis* and more energetically stable *trans*. In *trans* form, two chemical bonds attached to the central azo-group (N=N) are parallel to each other, so the molecule has an elongated (rod-like) form, while in the *cis* form the angle between these bonds is 120° and a molecule adopts a bent, more isotropic shape. In a polymeric matrix the *trans* molecules are randomly oriented and the system is optically isotropic before it is exposed to laser radiation. Due to different absorption spectra of the isomers any light induced variation of isomer concentration leads to the change of the absorption coefficient α and also the refractive index coefficient n of the film. When a molecule absorbs a photon and is excited, it comes back to the ground state by a nonradiative decay, either to the *trans* form or to the *cis* form. At room temperature, the *cis* form spontaneously relaxes towards the *trans* form, with a time constant depending on its environment (*i.e.* the nature of the chemical or physical links with the polymer). We assume that the number of DR1 molecules in a unitary volume is $N = N_T + N_C = \text{const}$ (*i.e.* there is no mass diffusion) and that $N_C \ll N$, which is valid for low light intensities (here C stands for *cis* and T for *trans*). Under steady state illumination condition, by light of energy suitable to convert *trans* to *cis* form of DR1 and back *cis* to *trans*, an equilibrium state between number of molecules in both forms is reached. The ratio of N_T/N_C is determined by the illumination level I, wavelength of light λ and various time constants of the molecular photoisomerization processes including thermal relaxation time constant τ. Absorbance of the film under steady-state light illumination condition reflects the ratio of N_T/N_C, because the absorption constants of both isomers are different $\sigma_T > \sigma_C$. For description of the optically induced anisotropy experiments in polymers containing chromophores an additional process must be taken into account, namely the "freezing" of the molecules in a polymeric matrix, the process that prevents them to undergo further photoisomerization. It is straightforward to assume that the number of molecules (N_F) not capable to undergo light induced conformational changes depends on the free space around a molecule, which is determined by the properties of polymer and dye-polymer specific interactions. So now we can write that $N = N_C + N_T + N_F$. All these quantities during steady-state illumination are time dependent, but their temporal evolutions are quite different. The kinetics of the N_C and N_T are closely related and can be determined by the rate equations given in [22], while the population of the "frozen" molecules undergoes a slow evolution with respect to the N_C and N_T. The situation becomes still more complicated if the excitation light is a monochromatic linearly polarized one. In such a case one deals with the angularly selective absorption. We assume that the absorption of *cis* isomer determined by the constant σ_C is isotropic, whereas that of *trans* isomer is dichroic with the molecular absorption tensor σ_T, having σ_T^z component much larger than $\sigma_T^x = \sigma_T^y$. For simplicity we assume that $\sigma_T^x = \sigma_T^y = 0$. For the linearly polarized light incident normally to the film surface, the system may be considered as having an axial symmetry. Now we can postulate the rate equations for the angular evolution of *cis, trans* and "frozen" isomers per unit volume N_C, $N_T(\Omega)$ and $N_F(\Omega)$:

$$\frac{\partial N_C}{\partial t} = -\sigma_C \beta_{CT} N_C I - \frac{N_C}{\tau} + N_T \int f(\nu(\Omega)) \beta_{TC} \cos^2 \theta I \, d\Omega \tag{2}$$

$$\frac{\partial N_T(\Omega)}{\partial t} = -\sigma_T \beta_{TC} N_T(\Omega) \cos^2 \theta I + \sigma_C \beta_{CT} N_C I \tag{3}$$

$$\frac{\partial N_F(\Omega)}{\partial t} = -\frac{N_F(\Omega)}{\tau_F} + N_T \int f(\nu(\Omega)) \beta_{TF} \cos^2 \theta I \, d\Omega \tag{4}$$

where $N_T(\Omega)$ and $N_F(\Omega)$ are the numbers of *trans* molecules oriented in the solid angle $d\Omega$ around the direction θ free to reorient (T) and ”frozen” not able to reorient (F); β_{CT} and β_{TC} are the quantum efficiencies of the photoisomerization of *cis* and *trans* isomers; β_{TF} describes quantum efficiency of the light induced molecular freezing process of *trans* isomers; $f(\nu(\Omega))$ is the distribution function of the long axis of the *trans* molecule; $d\Omega = \sin\theta d\theta d\phi$, where θ is a measure of the angle between the long molecular axis of the *trans* molecule and a plane determined by the light polarization; τ and τ_F are the time constants of the thermal relaxation of *cis-trans* and the relaxation of frozen *trans* molecules into free to reorient *trans* molecules. In the proposed set of differential equations we included the process of immobilization of *trans* molecules within the polymeric matrix, which disables them from reorientation. This process is connected with the matrix-chromophore interaction and the free volume around molecule that can change in time of illumination and strongly depends on temperature and matrix stiffness. The described process is much slower than the rotational diffusion process and faster than the thermal relaxation process for *cis-trans* transitions. For short illumination times this process can be neglected but for longer exposures it gives a quasi-permanent molecular alignment and must be taken into account in order to describe properly the experimental results. For short time of illumination (*i.e.* less than 1s), the condition that $N = N_T + N_C = \text{const}$ is a good approximation ($N_C << N$). However, for longer illumination times this condition should be replaced by $N = N_T + N_C + N_F = \text{const}$ ($N_F << N$). Since the sample shows important absorption at the working light wavelengths, we must introduce into our model the effects of the decreasing intensity of the laser light through the polymer film.

$$\frac{\partial I}{\partial z} = -I(A_1 N_C + A_2 N_T(\Omega, I)) \tag{5}$$

where A_1 and A_2 are the coefficients dependent on the wavelength. The set of coupled differential equations (2–4) cannot be solved analytically. However, the realistic approximations and simplifications made earlier [3,9,23,24] can give us an idea of the expected behavior of the systems under specific light illumination conditions. Knowing the temporal evolution of the angular distribution of *trans* molecules and the amount of *cis* molecules and "frozen" ones, the refractive index and absorption coefficient for linearly polarized laser beam can be calculated using a sophisticated Jones matrix formalism [25]. Because the holograms are written and measured in the self-diffraction experiment, then the coupled wave theory [25] must be used, including the variation of the chromophores orientation with thickness of the film and within the period of the light interference pattern. By a complex treatment and averaging over thickness the refractive index modulation created by the interference of writing beams for two orthogonal polarizations of incident beams along the y and the z axes could be represented by [25]:

$$\Delta n_z = \frac{n_{z,lighted} - n_{z,dark}}{2}; \qquad \Delta n_y = \frac{n_{y,lighted} - n_{y,dark}}{2} \tag{6}$$

where subscripts denote the index values in the lighted and dark zones of interference pattern. The diffraction efficiency of a thin hologram is given by $\eta = \sin^2\Delta\Phi$, where $\Delta\Phi$ is defined as $\Delta\Phi = (\pi d\Delta n_i)/\lambda$, where d is the hologram thickness and i = y or z.

Hologram recording at 632.8 nm light in polyimide – DR1-dye system in the presence of background illumination of 532 nm: Theoretical difficulties in calculation of the behavior of the sample under light illumination, especially in the case of hologram recording, led us to design a simple experiment, in which only one parameter is modified. We have prepared the experimental setup to measure the self-diffraction efficiency in the azo-dye doped polymer sample in the presence of another light illumination. By a proper choice of the intensity of the external light, not carrying any spatial modulation, we could study the influence of it on the recording mechanism itself. In Fig. 3 we present the preliminary self-diffraction results obtained in DR1 1% polyimide foil in the presence of YAG laser illumination. The plot of diffracted power *versus* time shows the dynamics of grating recording in the system. Note an increase of diffracted power on removal of YAG laser beam. The magnitude of the diffracted power increase depends on intensity of the 532 nm light illumination. This dependence was measured and is shown in Fig. 4, where the ratio of the diffraction signal increase ΔP_{diff} and the total diffraction power P_{total} *versus* intensity of YAG laser illumination are plotted. For about 4 mW/cm^2 intensity of 532 nm illumination a twofold increase of diffraction power is observed when this light is taken off. In this experiment the red light (He-Ne) intensity was I_0 = 343 mW/cm^2 *i.e.* in the bright fringe region the intensity was about 686 mW/cm^2. Calculating the photon flux for

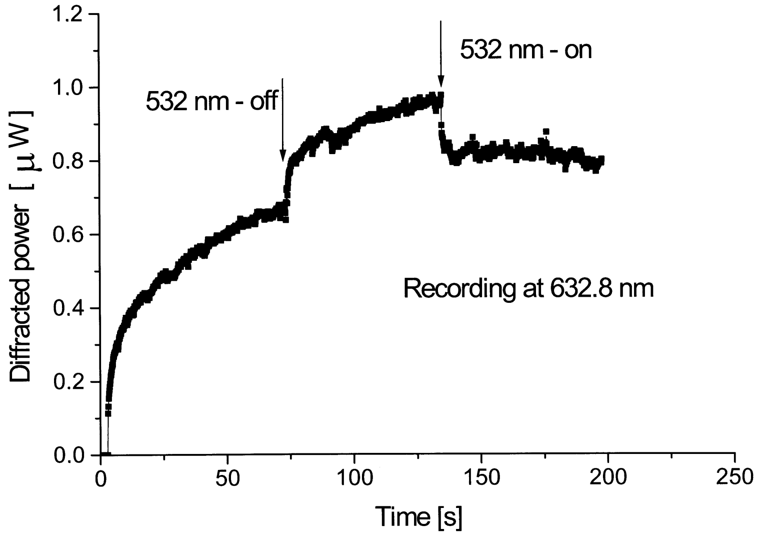


Figure 3. Evolution of diffraction power in function of exposure time in 1% DR1 doped polyimide foil (31 μm). Grating recording was realized for VV configuration of He-Ne laser beam polarizations (P_0 = 23.8 mW) in the presence of YAG laser beam (P = 380 μW) with V polarization. At 70 s the YAG laser beam was blocked for a period of 65 s and a clear rise of diffracted power occurred.

these two illumination conditions one arrives at $\Phi_{633} = 2.18\times10^{18}$ photons/cm^{2}s in the fringe center and $\Phi_{532} = 1.09\times10^{16}$ photons/cm^{2}s. Taking into account the difference in absorption coefficients of polyimide foil containing 1% of DR1 at $\lambda = 632.8$ nm $\alpha_{633} = 100$ cm^{-1} and at $\lambda = 532$ nm $\alpha_{532} = 2800$ cm^{-1}, one can estimate the number of absorbed photons/cm^{2}s within the 31 μm thick foil: $\Phi^a_{633} = 4\times10^{17}$ photons/cm^{2}s and $\Phi^a_{532} = 1\times10^{16}$ photons/cm^{2}s. This is a relatively small difference. Taking into account all the approximations, that we have made, one can conclude that the influence of the red and green light on the photoinduced molecular reorientation in the system studied is one order of magnitude higher for 532 nm light than for 633 nm light. We think that this reflects the resonant nature of the photoisomerization process.

Examining in detail the process of grating recording in the presence of YAG laser beam for VV configuration of He-Ne laser beam polarizations ($I_0 = 23.8$ mW) and V polarization of YAG laser beam ($I_0 = 380$ μW) shown in Fig. 3, we can determine the kinetics of grating recording. The power diffracted into the 1-st order as a function of time can be approximated by two-exponential function of the form:

$$P_{diff}(t) = A[1 - B\exp(-t/\tau_A) - C\exp(-t/\tau_B)] \tag{7}$$

where A is the saturation value of the diffracted power, B and C (B + C = 1) are the parameters describing the weighted contributions to the diffraction power coming from the two distinguishable processes. We link the faster process (B) with the equilibration of three processes: formation by light of *cis* states, formation by light of *trans* states and rotational diffusion of axially aligned molecules by the repeated two former processes. The slower process (C) we link with the equilibrium between the process of photoorientation of the *trans* molecules and a "freezing" of their position by the steric hindrance effect of the matrix and all other processes leading to relaxation of this ordering. The dynamics of the diffraction power shown in Fig. 3 can be fitted by:

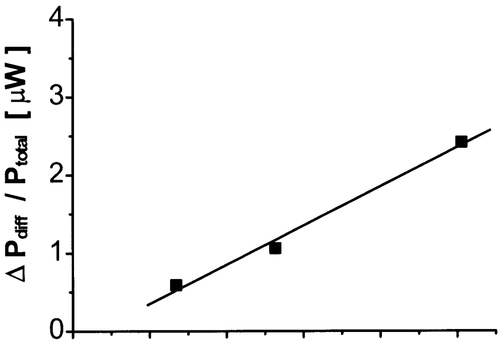


Figure 4. Plot of the ratio of diffraction power increase upon removal of 532 nm light (*cf.* Fig. 3) and the total diffracted power ($\Delta P_{diff}/P_{total}$) as the function of intensity of the YAG laser illumination.

$$P_{diff}(t) = 0.95[1 - 0.35\exp(-t/2.1) - 0.65\exp(-t/90)] \qquad (8)$$

where the time constants are expressed in seconds and diffracted power in μW, respectively. Subtraction of this function from the plot of P_{diff} (t) gives a net contribution to the diffraction power ΔP_{diff}(t) after taking off the YAG laser illumination. At the same time we subtract the slow component of grating buildup. The ΔP_{diff}(t) contribution is shown in detail in Fig. 5 and could be well fitted with two single exponential growth and decay functions:

$$\Delta P_{diff}(t) = 0.15[1 - \exp(-(t-70)/2.1] \quad \text{and} \quad \Delta P_{diff}(t) = 0.15\exp(-(t-135.5)/2.1) \qquad (9)$$

where the time constants are expressed in seconds and diffracted power in μW. This result is very interesting, as it tells us that the faster process of establishing the equilibrium value of photoinduced anisotropy under 632.8 nm illumination is described by the same time constant $\tau_B = 2.1$ s at the beginning of the grating recording ($t = 0$ s) as well as at the moment (here at $t = 70$ s), when the YAG laser is switched off. One should also notice that the relaxation time constant is also well described by the same value as grating increase time constant, *i.e.* 2.1 s. The measured by us time constants are not solely functions of material parameters as they also depend on experimental conditions like: beam intensities, temperature, geometry and light wavelengths.

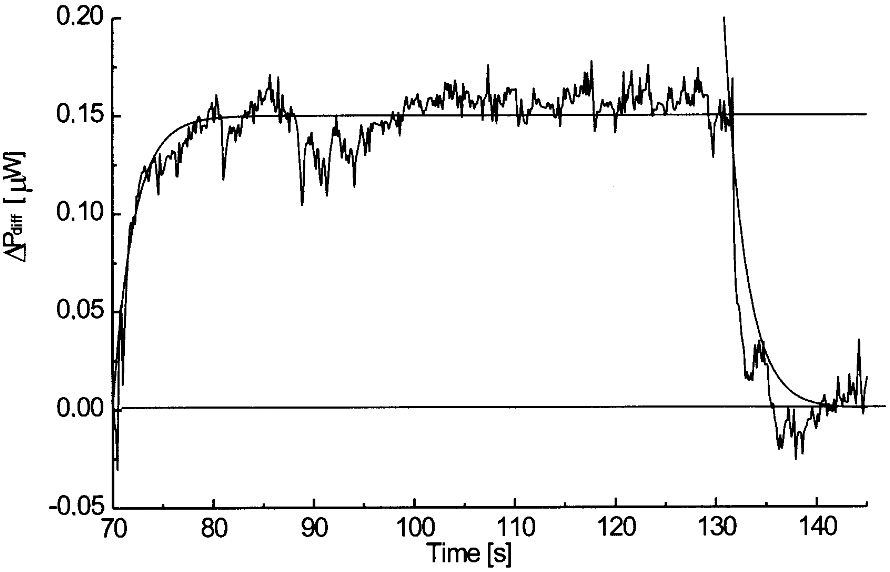


Figure 5. Part of the signal of diffracted power taken from Fig. 3 after subtracting the background signal in the time period 70–135 s. The curve is fitted with the single exponential growth and decay function with the same time constant $\tau_B = 2.1$ s.

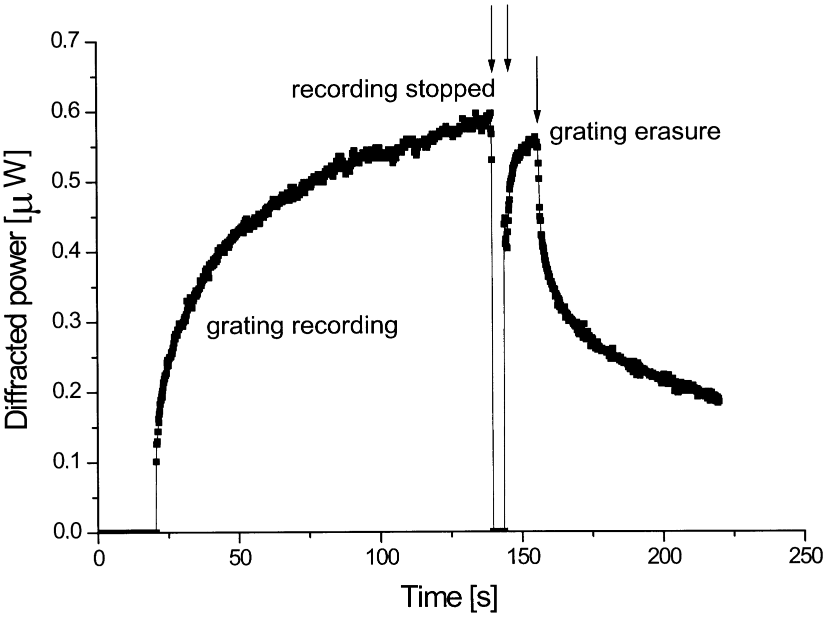


Figure 6. An example of evolution of grating recording by He-Ne laser light in DR1 1% polyimide. The recording was stopped at 140 s for 4.4 s (*i.e.* two beams were cut). Then again we recorded the grating for 12.5 s and next the one beam was taken off for the observation of light induced grating erasure.

In order to confirm that we really deal with the two distinguishable processes we present an experiment of grating recording by He-Ne laser light, which was stopped for few seconds by taking off the two writing beams. As one can see in Fig. 6, the subsequent reopening of the grating recording light beams leads to a sudden increase of diffraction power. This suggests that within the period of few seconds the signal drop was from the 0.58 μW to 0.38 μW, which is well understood on the basis of grating decay curve (when only one beams reads the grating) shown at the left part of the Fig. 6. The faster process is probably linked with buildup of population grating of excited *cis*-isomers and the slower one with appearance of the orientational grating due to ordering of *trans*-isomers.

Preliminary results of grating recording in AN doped polyimide system: The DR1 doped polyimide system was already studied by us [26]. In this paper we would like to present the preliminary results of grating recording in the novel system polyimide doped with the 1,3 diamino-4-azo(4′-nitroazobenzene)benzene (AN) shown in Fig. 1. This molecule with two N=N bonds probably also undergoes the photoisomerization process though its details can be different from that observed in DR1. We would like to check whether this molecule, when dissolved in the same polyimide matrix, could exhibit a photoinduced anisotropy like the DR1 does. The preliminary results are quite satisfactory as both intensity gratings (VV or HH configuration) and polarization gratings (VH configuration) could be recorded at 632.8 nm

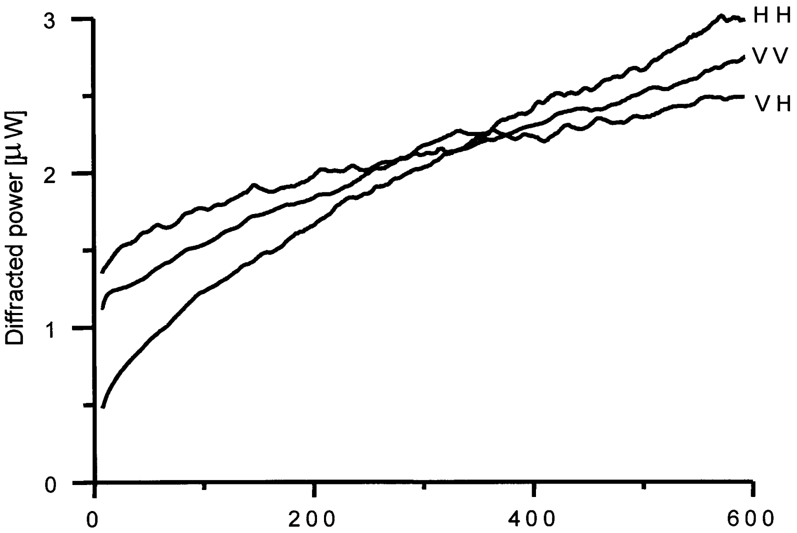


Figure 7. Evolution of the first order diffracted power as measured in two-wave mixing experiment with He-Ne laser light in 1% AN doped polyimide foil 31 μm thick. Symbols describe the polarization configuration of the beams pointing out that the polarization sensitive hologram recording (VH configuration) is possible in this system.

in the polymer foils (*cf.* Fig. 7). This strongly suggests that also this system allows for light induced molecular photoorientation. The dynamics of grating recording is also similar and composed of two processes faster and slower. The noticeable difference between the two systems is that in DR1 doped polyimide the VH configuration gives stronger grating for longer exposure and AN doped polyimide gives a weaker one. These preliminary results, however, need a much deeper analysis both experimental and theoretical. In Fig. 8 we present the experiment of grating recording by He-Ne light interrupted by opening of the YAG laser light incident on the grating area. There is also a well seen influence of a green 532 nm light on the diffraction efficiency, but the effect is much smaller than in the previous case. The differences between the two molecules are responsible for the effects observed. At present we could not offer any explanation, except the speculation that the two double N=N bonds in AN molecule in the presence of light leaves it in a more orientational freedom than DR1 molecule has in the same matrix.

Conclusions: Polymers doped with dichroic photoisomerizable molecules enable the recording of optical information through optically induced photoanisotropy. This ability extends through the whole absorption band of the dye, thus enabling multicolor holography. The unique property of the polymer systems containing azo-dyes, like the well-known DR1, is that these materials may record also polarization sensitive holograms. These features are possible, due to the mechanism of the photoinduced molecular reorientation of dichroic molecules in the polymeric matrices. Despite the hundreds of papers devoted to these materials, still the microscopic

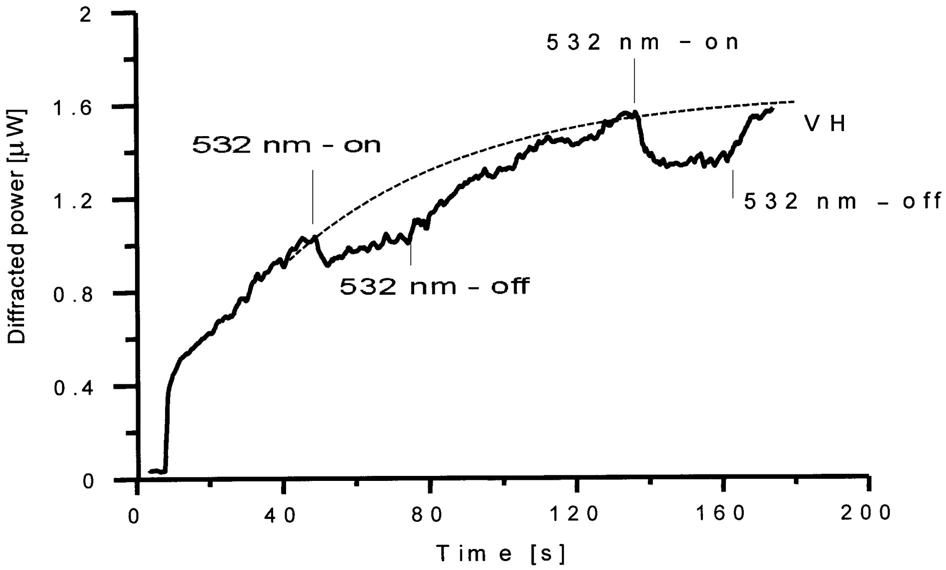


Figure 8. Self-diffraction dynamics measured in AN doped polyimide foil in the presence of YAG laser illumination. He-Ne laser light power was 25.8 mW and the beams were VH polarization, YAG laser had the power of 3.1 mW and the beam has V polarization.

mechanisms linked with diffraction grating formation are studied as they depend not only on the dissolved molecules but also on the properties of the matrix. In this paper we report on the influence of light on *in situ* recorded hologram in high T_g polyimide containing DR1 molecule and newly synthesized 1,3 diamino-4-azo(4′-nitroazobenzene)benzene. The investigations were aimed at search of some novel polymeric systems for dynamic holography. In the domain of optoelectronics and photonics there is a need for dynamic multicolor steering of optical properties of materials used in integrated optical circuits.

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