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UV-laser induced birefringence in oligoetheracrylate photopolymers

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ABSTRACT

Laser induced birefringence was studied during coherent treatment by two coherent UV laser beams at wavelength 355 nm. Principal role of the border separating photo-solidified and the surrounding non-solidified liquid-like fragments is demonstrated. The magnitudes of laser induced birefringence for probing 1150 nm wavelength were varied within $10^{-4} \dots 10^{-2}$. It is demonstrated that one can operate by values of the birefringence versus polymer solidification time. The UV-induced birefringence achieves a saturation after about 45 days. The reversible birefringence after switching off of the UV-solidification beams was equal to about 18% with respect to initially photo-stimulated. The discovered effect may be used for design and fabrication of electro-optical modulators operated by UV laser light. A principal physical mechanism is determined by an interaction between two coherent UV- nanosecond laser pulses at wavelengths of 355 nm (third harmonic of Nd:YAG laser). The discovered effects allow to propose a new type of UV-operated photopolymer materials for optical recoding of information.

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1. Introduction

Liquid oligoether acrylate (OEA) photopolymer composites are solidified by UV laser light. Generally there is applied single laser beam. Due to different time of solidification process there occur some layers which possess different refractive indices and mechanical properties [1–3]. At the same time the use of two coherent UV laser beams may open a new technology for formation of the optical anisotropy. This anisotropy is directly related to optical birefringence. This one may be promising for design and production of UV-induced anisotropic materials [4]. Such effects may be used for dynamical holography, optical memory, triggering etc. Very crucial here is an existence of different local space non homogenous fragments [5]. Additionally there occur some intermediate phases which are quite similar to liquid crystalline phases. Such phases can give substantial contribution to optical susceptibilities including nonlinear optical and electro-optical ones [6]. Generally the effect is similar to anisotropy which is occurred near liquid crystalline phase borders [7]. To achieve a more efficient photoinduced optical anisotropy additional illumination was performed using two coherent UV-laser beams. These beams form a space grating similarly to the described in Ref. [8]. The main effect is a consequence of interference of interacting coherent beams incident at different angles. Up today the

formation of the photoinduced optical anisotropy was realized for different states of condensed matter [9], metallic nanoparticles [10], $\text{Ge}_3\text{Sb}_2\text{Te}_6$ ferroelectrics [11] etc. Particular interest present polymer composites with embedded oxide materials [12]. The use of such kind of materials gives an opportunity to achieve a high degree of operation by optical parameters using a relatively facile technology [13]. Very important here is to form the composites with solidified granular sizes varying within $10 \dots 30 \mu\text{m}$ [14]. The interface regions [15] play here crucial role.

In the present work we will create the tunable optical anisotropy using the different time of UV-solidification. Very important parameter here is optical power density photoinduced beams which defines principal changes of the optical constants (absorption and refractive indices) [16]. The prevailing mechanisms here are optical Kerr effect described by fourth rank tensors [17], excitation of free carriers [18], photo-polarization [19], non-linear light scattering. However, the main restraining factor here is an existence of parasitic light scattering during formation of phases possessing different refractive indices. The coexistence of photoinduced linear and nonlinear optical constants may be very efficient factor for production of optically operated devices.

As a consequence, in the present work, we will perform two-beam coherent laser induced tuning of optical anisotropy (controlled by birefringence) versus the UV-solidification time. The effect will be explored using the microscopical images of solidified samples as well as direct monitoring of the birefringence.

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2. Methodology

The general procedure of the oligoetheracrylate (OEA) photopolymers synthesis was similar to the described in Ref. [20]. The principal schema of the UV-induced photo-solidification and control of anisotropy is presented in Fig. 1.

The liquid photopolymer is put into the thermo regulated thermo-chamber varying temperature within 0 °C...120 °C with thermo-stabilization up to 1 °C. The thermo-chamber is supplied with quartz windows. The UV cw laser generating at 371 nm with power density 45 W/cm² was used for performance of OEA solidification. Its deviation from the average power (stability) did not exceed 0.25%. The beam diameter was equal to about 6 mm and space profile was close to Gaussian like. The beam intensity stability was better than 10⁻². A special camera CCD connected with the PC was used for layer-by-layer tomography monitoring of the UV-solidification process in real time. The process of solidification was performed from 1 min. up to 5 min and was controlled by cw He-Ne laser at 1150 nm wavelength with light power equal to about 15 mW. A Glann prism rotating analyzer A was used for control of the birefringence with accuracy up to 5 * 10⁻⁵. Generally the Senarmont method is similar to the described in Ref. [21]. The probing beam was completely covered by the photoinducing beam and this exceed was about 12%.

The photoinduced two-beam coherent splitting was performed using the set of mirrors M1, M2, M3, M4, (see Fig. 1). Frequency tripled beam of fundamental Nd: YAG laser was incident on the samples and was split for two coherent beams which incident to solidified samples at angles varying within the 12...24 degree. Their beam profiles were of Gaussian like form with degree 0.15. The system of phase inverter plates allowed to vary ratio of intensities between the two coherent beams at 355 nm.

The typical photo induced treatment process is varied versus the photo solidification time. Following Fig. 2 one can see that during solidification there occur some interface borders. Each layer possesses different refractive index and on the borders there occur some space gradients of refractive indices. The interfaces between the layers are varied from 20 nm up to 100 nm. These gradients play principal role for the magnitudes of ground state dipole

moments which define the corresponding nonlinear optical hyper-polarizabilities [22]. Depending on the time of UV illumination these space gradients are varied. The performed evaluations have shown that these space gradient's contribution are up to 7%. During the first few minutes of the UV illumination the solidification process is expanded with different rate and after 5 min. the solidified phase becomes dominant. The light scattering background was monitored and its value was below 2...3%. The monitoring of the interfaces was done by direct observation of the photoinduced coherent gratings. More details will be given below.

3. Results and discussion

In Figs. 3–5 are shown computationally generated images of the transparency versus the time of UV-solidified illumination. It is clearly demonstrated that the shape and sizes of the corresponding optically polarized transparency are crucially dependent on the illumination time. It may also reflect different space topology of the interfaces versus the time of UV solidification. The observed birefringence presents a statistically superimposed anisotropy originated from different parts of the photo-solidified grains. The process is observed only during the UV-solidification process. Afterwards there occurs some saturation of the photoinduced birefringence.

Following the presented pictures one can clearly see different regions forming the charge density space acentricity. This factor favors different space profiles of the birefringence.

The polarization of the two coherent photoinduced beams had not significant influence on the observed images. This fact may confirm a complete disordering of the particular photo-solidified grains. Additional source of the observed pictures may be Kerr effect contribution described by fourth rank tensors.

The photoinduced Kerr effect may be also dependent on the number of trapping levels on the borders [23] and the free carrier excitations. However after switching off of the two-beam UV coherent treatment the effect disappears. This one additionally confirm the principal role of the gratings in the effect. It is crucial that Figs. 3 and 4 are very similar what indicates that during the

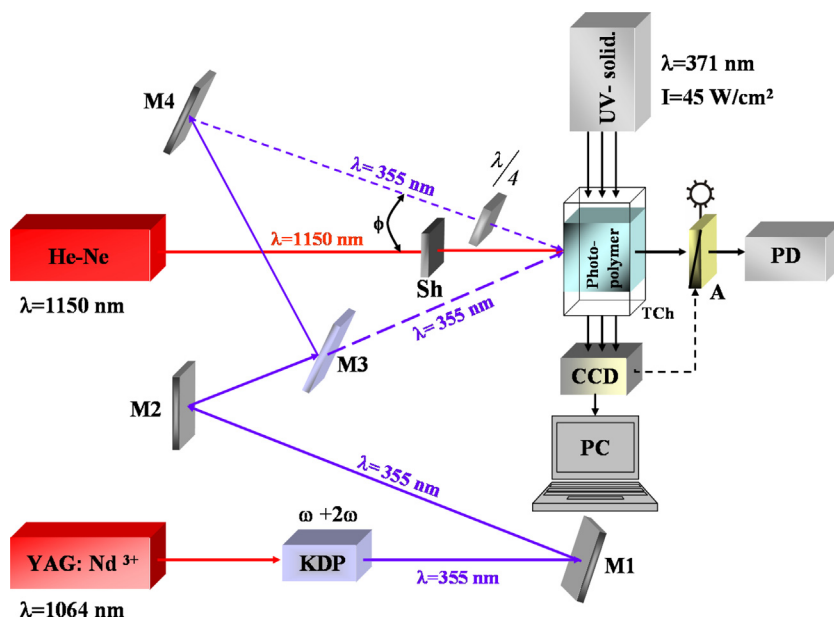


Fig. 1. Set-up for photoinduced treatment of photopolymers: YAG: Nd³⁺ –1064 nm 20 ns laser at $\lambda = 1064$ nm with frequency repetition 5 Hz; KDP – the nonlinear optical crystal creating third harmonic generation with frequency $\omega+2\omega$ summation; M1, M2, M4 – a set of mirrors. The M3 is semi-transparent mirror. The processes of the UV-solidification, two-beam coherent treatment and birefringence control are timely synchronized and the process is controlled by PC connected with CCD camera.

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