

Multiple cycles of recording/reading/erasing birefringence in azopolymers

L. Nedelchev^{1,2,*}, D. Ivanov¹, G. Mateev¹, B. Blagoeva¹, D. Kostadinova¹,
N. Berberova¹ and D. Nazarova¹

¹Institute for Optical Materials and Technologies "Acad. J. Malinowski", Bulgarian Academy of Sciences,
"Acad. G. Bonchev" str. Bl. 109, 1113 Sofia, Bulgaria

²University of Telecommunications and Post, Acad. St. Mladenov 1 Str., Sofia 1700, Bulgaria

Received: October 10, 2016; Revised: November 10, 2016

In this work we investigate the characteristics of reversible polarization recording in an azopolymer. Polarized light from DPSS laser with wavelength 444 nm is used for recording photoinduced birefringence. Afterwards, two methods for its erasure are used: heating the polymer to certain temperature or illumination with circularly polarized light. Multiple cycles of recording and erasure are successfully realized with both methods, which allows us to compare and outline the advantages of each of them. The photoinduced birefringence is calculated from the Stocks parameters, monitored in real time, during the entire experiment.

Keywords: Azopolymers; Thermal and optical reversibility; Photoinduced birefringence; Polarization recording.

INTRODUCTION

In contrast to conventional optical data storage (CD/DVD/Blu-ray technologies), where recording is in one plane (2D), holographic data storage allows to address the entire volume of the media (3D) and to achieve much higher density of information [1, 2]. Polarization holography makes possible to increase the information density even further by registering also the polarization state of the recording light and realizing polarization multiplexing [3-5].

Since 1984, when Todorov et al. [6] discovered the possibility to inscribe high-efficient polarization holograms in azobenzene-containing materials, these materials are subject of intensive research for numerous application, including holographic data storage [7, 12]. Essential parameters for each data storage media are the stability of the recorded information, as well as its reversibility i.e. the ability of the media to ensure multiple cycles of recording, non-destructive reading and erasure of information.

Here we present a study of the reversibility properties of birefringence in amorphous azo polymer. The value of the photoinduced linear birefringence Δn is defined as the difference between the component of the refractive index perpendicular to the polarization of the recording

beam and the component parallel to it – $\Delta n = n_{\perp} - n_{\parallel}$. It is known that on illumination with linearly polarized light within the absorbance band of the photochromic azo groups, they undergo cycles of *trans-cis-trans* isomerizations and tend to reorient perpendicularly to the polarization direction of light [3, 6]. As a result, birefringence is induced in the material at a macroscopic level. On the other hand, when the azopolymer film is illuminated with circularly polarized light, no preferential direction exists, so the molecules orientation becomes random again, and the birefringence is erased.

We apply two methods of erasure in this study – optical and thermal. Both methods are compared in terms of speed of erasure and anisotropy values retention during multiple write/read/erase cycles.

EXPERIMENTAL

Materials

The polymer used in this experiment is side-chain amorphous azocopolymer, denoted as P₁₋₂, synthesized earlier in the Institute of Optical Materials and Technologies [13]. The azopolymer chemical structure and spectrum of absorbance are shown in Fig. 1. Its molecular weight is $M_w = 3600$ g/mol and glass transition temperature is $T_g = 102$ °C [14].

* To whom all correspondence should be sent:
E-mail: lian@iomt.bas.bg

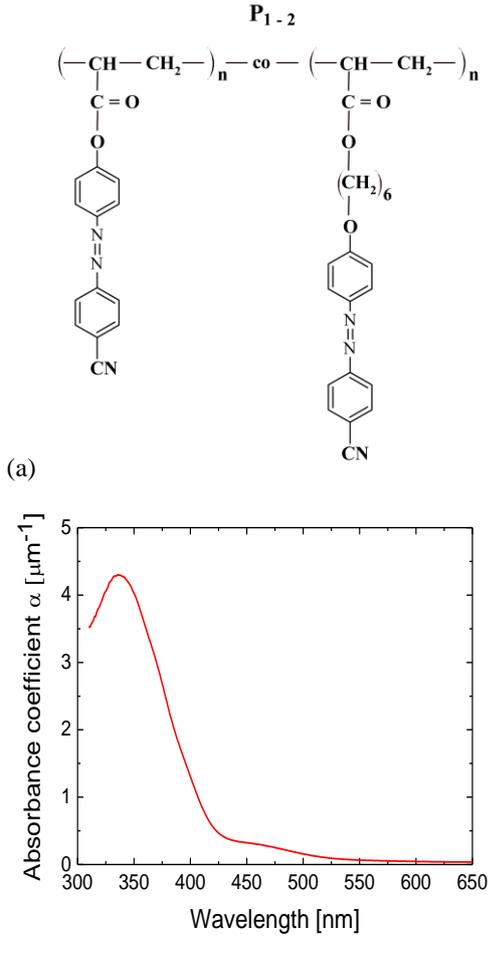


Fig. 1. (a) Chemical structure of the azocopolymer P₁₋₂; (b) Spectrum of absorbance of P₁₋₂.

Methods of characterization

The preparation of the polymer films includes dissolving of the P₁₋₂ copolymer in 1,2-dichloroethane and spin coating the solution at 1500 rpm on glass substrates. As a result, thin homogeneous films are formed. The thickness of the films is measured by a Talystep profilometer (Taylor Hobson) and for the samples used in this work is $d = 700$ nm.

To record and probe the photoinduced birefringence, two diode-pumped solid state lasers (DPSSL) are used: recording laser with $\lambda_{rec} = 444$ nm, power 35 mW, and reading (referred also as probe) laser with $\lambda_{probe} = 635$ nm and power below 2 mW. As it can be seen in Fig. 2, the reading laser beam passes through polarizer, oriented at 45°, while the recording beam passes through vertical polarizer. After passing through the azopolymer sample, the reading beam enters a polarimeter (PAX5710 Polarization Analyzing System, Thorlabs), which monitors the Stokes parameters of light in real time.

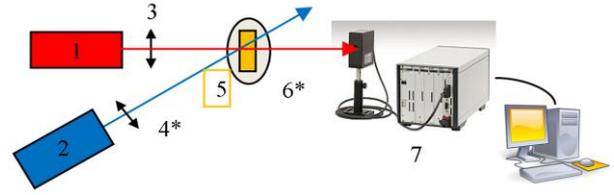


Fig. 2. Experimental setup : 1 – reading laser (635 nm, $P < 2$ mW), 2 – recording laser (444 nm, $P = 35$ mW), 3 – polarizer oriented at 45°, 4* – vertical polarizer/QWP, 5 – azopolymer sample, 6* – hot stage, 7 – Thorlabs polarimeter. (* denotes an element, which is replaceable, depending on the erasing method used).

In this case the birefringence can be calculated from the expression:

$$\Delta n = \frac{\lambda_{probe}}{2\pi d} \arctan\left(\frac{S_3}{S_2}\right) \quad (1)$$

where d is the film thickness, and S_2 and S_3 are Stokes parameters [10, 15].

RESULTS AND DISCUSSIONS

Fig. 2 is a representation of the experimental setup for both deleting methods. The reading laser is on throughout the entire experiment. Initially, 60 seconds of background signal is measured, without illumination with the recording laser. During the data processing, these background values are subtracted from the measured data. Then we start the illumination with the recording laser until the photoinduced birefringence reaches saturation – Δn_{max} . Using Eq. (1), the time dependence of the birefringence can be determined.

Optical erasure

During the recording, the light from the laser at 444 nm passes through a vertical polarizer, while for the erasure it is replaced by quarter wave plate (QWP), which converts the linearly polarized light in circularly polarized (CP). CP light with the same wavelength as the recording one, randomizes the orientation of the azochromophores which leads to erasure of the birefringence.

The processes of recording and deleting in the azopolymer film had been repeated 10 times, all in the same place of the sample. The results are shown in Fig. 3. The recording time for each cycle is 4 minutes, then the recording laser is stopped and only the reading beam is on for another 4 minutes to determine the stability on reading. After that, starts the illumination with circularly polarized light at 444 nm for another 4 minutes.

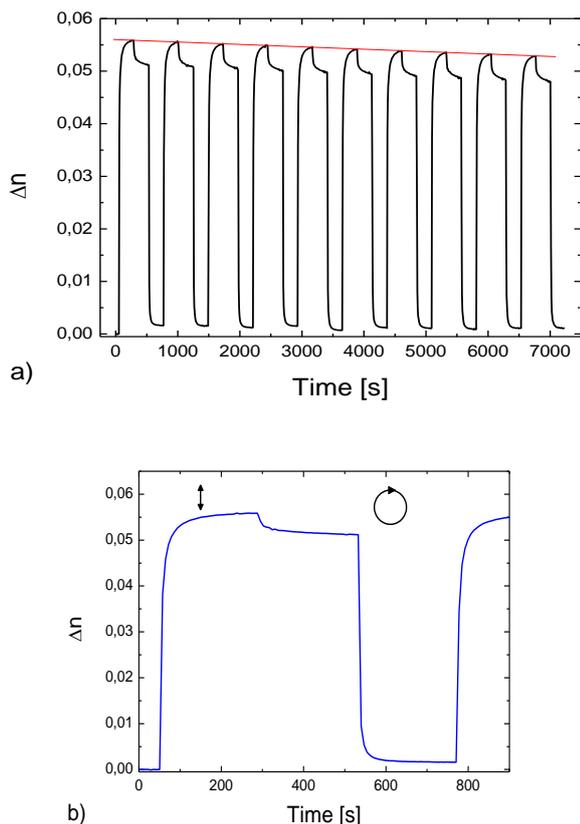


Fig. 3. (a) Ten cycles of inducing and optically deleting linear birefringence; (b) Zoom on the first cycle. The symbols indicate the state of polarization of the laser with $\lambda = 444$ nm.

As can be seen from Fig. 3, this polymer can be successfully used for multiple cycles of recording and deleting of linear birefringence. The response of the polymer to different polarizations is fast and relatively high values of birefringence have been achieved. However, we should note, that after each cycle the maximum value of birefringence is reduced. This phenomena could be explained with the reorientation of part of the chromophores perpendicular to the film surface [9]. Hence, these molecules do not contribute to the formation of birefringence.

Thermal erasure

To study the thermal erasure, the sample was mounted on a hot stage THMS600 (Linkam Scientific), which can maintain or vary sample temperature at a given speed, with high precision. The recording and relaxation time in this experiment were set to 2 minutes. After that time, starts the heating process with rate $10^{\circ}\text{C}/\text{min}$. The starting temperature for the measurement was 30°C . The heating continues until temperature of 90°C is reached, which leads to erasure of the photoinduced

birefringence, as shown in Fig. 4. Then the temperature is decreased back to 30°C for the next cycle of recording/reading/erasure. It is important to notice, that in this case no decrease of the maximal value of birefringence in the consecutive cycles is observed.

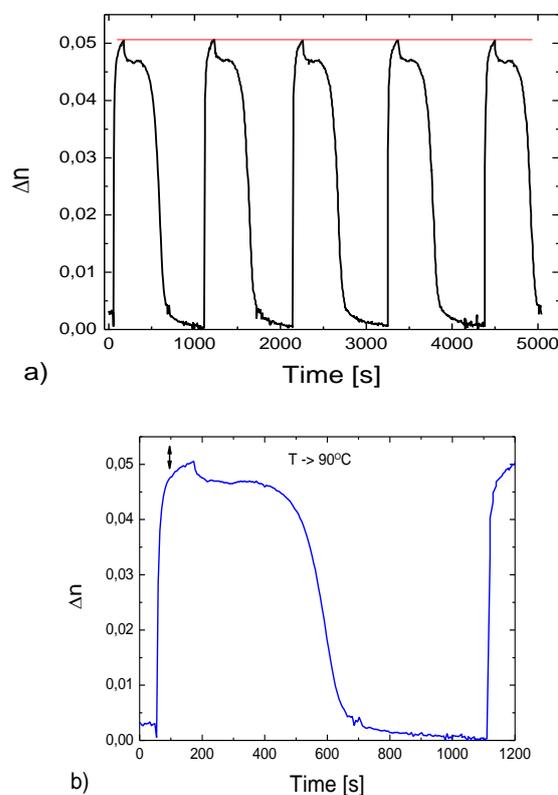


Fig. 4. (a) Five cycles of inducing and thermally deleting linear birefringence; (b) Zoom on the first cycle. The arrow indicates the polarization state of the pump laser.

CONCLUSIONS

We have demonstrated experimentally the possibility for multiple cycles of recording, reading and thermal or optical erasure of photoinduced birefringence in the azocopolymer P_{1-2} synthesized in IOMT. Reversible recording with linearly polarized light and erasure with circularly polarized light at the same wavelength is realized, in contrast to other methods where a separate UV laser is used for the erasure [9]. The processes can be repeated many times only with slight decrease of the amplitude – less than 5% for 10 cycles. A possible reason for this behaviour can be the fact that some molecules reorient perpendicularly to the plane of the circularly polarized light. Being in such position they cannot be addressed by the linearly polarized light in the next recording cycle, which results in decrease of the maximal value of the birefringence (Δn_{max}), as seen in Fig. 3a. This drawback has been

overcome using thermal erasure. By this method the anisotropy can be also easily erased by heating to temperature of around 90°C and the films can be used for recording again. Although the thermal erasure is slower, we have shown that it retains the maximal value of birefringence, as indicated by the red line in Fig. 4a. Therefore, the erasure method can be selected depending on the application, or alternatively, after a given number of cycles using optical erasure, a thermal erasure can be applied in order to restore the birefringence value to its maximal level.

Acknowledgements: Authors are grateful for the financial support provided by Bulgarian Science Fund under the project DCOST 01/7 and also acknowledge the COST Action MP1205.

REFERENCES

1. S. Orlov, W. Phillips, E. Bjornson, Y. Takashima, P. Sundaram, L. Hesselink, R. Okas, D. Kwan, R. Snyder, *Appl. Opt.*, **43**, 4902 (2004).
2. K. Curtis, L. Dhar, A. J. Hill, W. L. Wilson, M. R. Ayres, *Holographic Data Storage: From Theory to Practical Systems*, John Wiley & Sons, New Jersey, 2010.
3. L. Nikolova, P. Ramanujam, *Polarization Holography*, Cambridge University Press, Cambridge, 2009.
4. L. Nedelchev, D. Ilieva, Tz. Petrova, N. Tomova, V. Dragostinova, L. Nikolova, *Proc. SPIE*, **5830**, 95 (2005).
5. D. Ilieva, L. Nedelchev, Ts. Petrova, N. Tomova, V. Dragostinova, L. Nikolova, *J. Opt. A: Pure Appl. Opt.*, **7**, 35 (2005).
6. T. Todorov, L. Nikolova, N. Tomova, *Appl. Opt.*, **23**, 4309 (1984).
7. A. Natansohn, P. Rochon, J. Gosselin, S. Xie, *Macromolecules*, **25**, 2268 (1992).
8. S. Hvilsted, F. Andruzzi, C. Kulinna, H. W. Siesler, P. S. Ramanujam, *Macromolecules*, **28**, 2172 (1995).
9. N. C. R. Holme, P. S. Ramanujam, S. Hvilsted, *Opt. Lett.*, **21**, 902 (1996).
10. L. Nedelchev, A.S. Matharu, S. Hvilsted, P.S. Ramanujam, *Appl. Opt.*, **42**, 5918 (2003).
11. H. Audorff, K. Kreger, R. Walker, D. Haarer, L. Kador, H.W. Schmidt, *Adv. Polym. Sci.*, **228**, 9 (2010).
12. A. Ryabchun, A. Sobolewska, A. Bobrovsky, V. Shibaev, J. Stumpe, *J. Polymer Sci., Part B: Polym. Phys.*, **52**, 773 (2014).
13. G. Martinez-Ponce, T. Petrova, N. Tomova, V. Dragostinova, T. Todorov, L. Nikolova, *J. Opt. A: Pure Appl. Opt.*, **6**, 324 (2004).
14. L. Nedelchev, D. Nazarova, V. Dragostinova, *J. Photochem. Photobiol. A: Chem.*, **261**, 26 (2013).
15. N. Berberova, D. Nazarova, L. Nedelchev, B. Blagoeva, D. Kostadinova, V. Marinova, E. Stoykova, *J. Phys.: Conf. Series*, **700**, 012032 (2016).

МНОГОКРАТЕН ЗАПИС/ЧЕТЕНЕ/ИЗТРИВАНЕ НА ДВУЛЪЧЕПРЕЧУПВАНЕ В АЗОПОЛИМЕРИ

Л. Неделчев^{1,2}, Д. Иванов¹, Г. Матеев¹, Б. Благоева¹, Д. Костадинова¹, Н. Берберова¹ и Д. Назърова¹

¹Институт по оптически материали и технологии "Акад. Й. Малиновски", Българска Академия на науките, ул. "Акад. Г. Бончев", бл. 109, 1113 София, България

²Висше училище по телекомуникации и пощи, ул. «Акад. Ст. Младенов» 1, 1700, София, България

Постъпила на 10 октомври 2016 г.; коригирана на 10 ноември 2016 г.

(Резюме)

Изследван е процесът на многократен обратим поляризационен запис в азополимер. За запис на фотоиндуцирано двулъчепречупване е използвана поляризирана светлина от лазер с дължина на вълната 444 nm. За изтриване на записа са използвани два метода: загряване на полимерния слой или облъчване с кръгово поляризирана светлина. Успешно са реализирани многократни цикли на запис и изтриване и чрез двата метода, което ни позволява да ги сравним и посочим преимуществата на всеки от тях. Фотоиндуцираното двулъчепречупване е изчислено от параметрите на Стокс, които се следят в реално време по време на целия експеримент.