This article was downloaded by: [University of Haifa Library]
On: 20 August 2012, At: 20:20
Publisher: Taylor & Francis
Informa Ltd Registered in England and Wales Registered Number: 1072954
Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

Study of Optical Switching and Reorientation in Liquid Crystals of Homologous Series of 4-n-butyl-4′-n-alkoxyazobenzenes

Vladimir Grozhik [a] , Helen Gleeson [b] , Svetlana Serak [c] , Samanta Watson [b] & Alexander Agashkov [c]

[a] Institute of General and Inorganic Chemistry of NASB, 9, Surganova, Minsk, 220072, Belarus

[b] Schuster Laboratory, Department of Physics and Astronomy, University of Manchester, Manchester, M13 9PL

[c] Institute of Electronics of NASB, 22, Lagoiski Trakt, Minsk, 220090, Belarus

Version of record first published: 24 Sep 2006

To cite this article: Vladimir Grozhik, Helen Gleeson, Svetlana Serak, Samanta Watson & Alexander Agashkov (1998): Study of Optical Switching and Reorientation in Liquid Crystals of Homologous Series of 4-n-butyl-4′-n-alkoxyazobenzenes, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 320:1, 433-444

To link to this article: http://dx.doi.org/10.1080/10587259808024413

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: http://www.tandfonline.com/page/terms-and-conditions

This article may be used for research, teaching, and private study purposes. Any substantial or systematic reproduction, redistribution, reselling, loan, sub-licensing, systematic supply, or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae, and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand, or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

Downloaded by [University of Haifa Library] at 20:20 20 August 2012

Mol. Cryst. Liq. Cryst., Vol. 320, pp. 433–444
Reprints available directly from the publisher
Photocopying permitted by license only

© 1998 OPA (Overseas Publishers Association) N.V.
Published by license under
the Gordon and Breach Science
Publishers imprint.
Printed in Malaysia.

Study of Optical Switching and Reorientation in Liquid Crystals of Homologous Series of 4-n-butyl-4'-n-alkoxyazobenzenes

VLADIMIR GROZHIK [a], HELEN GLEESON [b], SVETLANA SERAK [c], SAMANTA WATSON [b] and ALEXANDER AGASHKOV [c]

[a] Institute of General and Inorganic Chemistry of NASB, 9,Surganova, Minsk, 220072, Belarus; [b] Schuster Laboratory, Department of Physics and Astronomy, University of Manchester, Manchester M13 9PL; [c] Institute of Electronics of NASB, 22, Lagoiski Trakt, Minsk, 220090, Belarus

Downloaded by [University of Haifa Library] at 20:20 20 August 2012

Two mechanisms of optical response in liquid crystals (LC) of homologous series 4-n-butyl-4'-n-alkoxyazobenzenes (BAAB), namely the photochemical phase transition and reorientation in bulk of LC are considered. The temporal parameters of writing (Ar^+ laser) and erasing (He-Ne laser) of information on BAAB on the basis of photoinduced phase transition are presented. The phenomenon of reorientation of BAAB molecules in planar oriented layer induced with second of Nd:YAG laser in directions orthogonal to initial alignment was revealed.

Keywords: azobenzenes; liquid crystals; phase transition; reorientation; laser

INTRODUCTION

At present time as a media for optical registration of information are widely used LC, polymeric liquid crystals (PLC), dye doped liquid crystals (DDLC), molecules of which have some structural fragments that are able to its geometry change under the action radiation from lasers. Registration of information by such substrates can be based on various mechanisms, they are, for example, the photochemical phase transition into isotropic state[1-6], the change of parameters of a selective reflection band of chiral LC's[7], the change

of polarization in ferroelectric LC's[8], the different modes of director reorientation[9-17], but all of them are joined with photoisomerization of substrate molecules. Photoinduced transition of double chemical π-bonds in rod-like molecules to single σ-ones gives the possibility of free rotation of molecular fragments around σ-bond and transition of molecules into thermodynamically unstable cis-isomers in such compounds as azomethynes, stilbenes and azocompounds. Obtained cis-isomers can relax back into trans-isomers within a quite wide scale of times (from a few μs in case of azomethynes to hours in case azocompounds[18]). For the registration of information the most interesting are compounds having long-living cis-isomers. As a such compounds the LC's of homologous series of 4-n-butyl-4'-n-alkoxyazobenzenes (BAAB) can be used.

This report presents the results of investigation of parameters of optical response, induced in BAAB with radiation of the laser. In these compounds under the action of radiation first of all one can be observed the lowering of phase transition temperature T_c that is well-known phenomenon in the mixtures of individual components, in our case of trans- and cis-isomers. On the other hand changes in molecules geometry and their polarizability at photoisomerization can induce to change of initial orientation of LC-layer. Gibbons et al.[9] reported about registration of information as a result photoinduced reorientation of azopolymer coating, that in turn caused twist-reorientation LC director. By using of various polymer coating, doped with azobenzenes, one can reorient the layer director from homeotropic state to planar one[11] or vice versa[12]. Authors[13] investigated photoinduced reorientation in bulk of the azodyes doped LC (1% by weight). This reorientation in some DDLC can be orthogonal to polarization vector of exciting wave and to director of its propagating (molecules turn in plane of substrates of planar cell), in other cases such reorientation can coincide with the wave vector of wave propagation (normally to substrates). Also it is possible to observe the third case that is rotation of LC molecules in either of two directions, each of them is orthogonal to initial alignment. Rotation of layer director in bull of DDLC can cause the appearance of easy axis on unrubbing substrate of planar cell[16-17].

Downloaded by [University of Haifa Library] at 20:20 20 August 2012

We investigated two mechanisms of interaction in LC-azobenzenes, that are connected both with photochemical phase transition and with reorientation in BAAB.

EXPERIMENT AND RESULTS

1.Photoinduced phase transition in BAAB.

The method of registration of information in LC based on photoinduced phase transition was for the first time reported in work of Haas et al.[1]. They were radiating the mixture of stilbenes in nematic phase with Xe-Hg lamp (λ=300 nm). Analogous method of optical information registration we realized[2] on mixture of LC-azobenzenes at interaction of them with radiation from He-Cd laser (λ=440 nm, P=20mW) and from ruby laser (second harmonics, λ=347nm, Δt=50ns, W=20 mJ/cm^2). The spatial resolution in that experiment was at least of 25 lines/mm in planar LC-cell at the thickness of 20μm. Fig. 1 shows photo of the image of photomask, focused on LC-layer by lens. At use of He-Cd laser the exposure time was 2-3s (d=20μm). Ruby laser formed the image by one pulse. Authors[6] showed that cis-isomer forms in azobenzenes in very short time scale, about 10 ns.

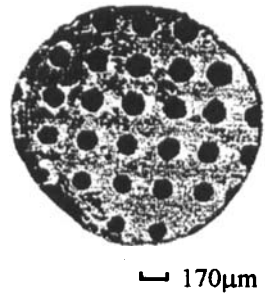


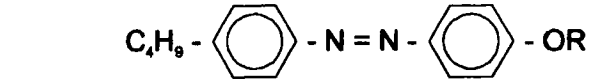
FIGURE 1 Photo of the stored image in BAAB

We carried out the investigation of times of writing τ_{wr} and erasing τ_{er} in eight homologs of 4-n-butyl-4'-n-alkoxyazobenzenes and in the mixture of that homologs C1:C2:C5 (1:1:1 by weight) (BAAB-1), having nematic phase at the room temperature. The compounds were synthesized by the method, described in [19]. Their structure formulas, phase transition points and dependences of refractive indexes n_o, n_e vs. temperature are shown in Fig. 2.

Planar LC-cell was irradiated by Ar^+ laser (λ=488 nm, P=50 mW/cm^2), polarization vector of beam coincided with layer director L. Registration of written information was carrying out on input glass of cell that was set between crossed polaroid's by the He-Ne laser (λ=633nm, P=50mW/cm^2). The angle between layer director and optical axis of one of polaroid's was equal to 45°.

Downloaded by [University of Haifa Library] at 20:20 20 August 2012

Planar alignment of LC molecules was achieved by treatment of glass surfaces with polyimide. The cell was placed in heating chamber with Linkam TMS90



R=C_2H_5 K48N83I
R=C_3H_7 K67N68I
R=C_4H_9 K65N75I
R=C_6H_{13} K44SmA(41)N74I
R=C_7H_{15} K56SmA(55)N72I
R=C_8H_{17} K57SmA(58)N77I
R=C_9H_{19} K63SmA(64)N74I
R=$C_{10}H_{21}$ K55SmA(71)N76I

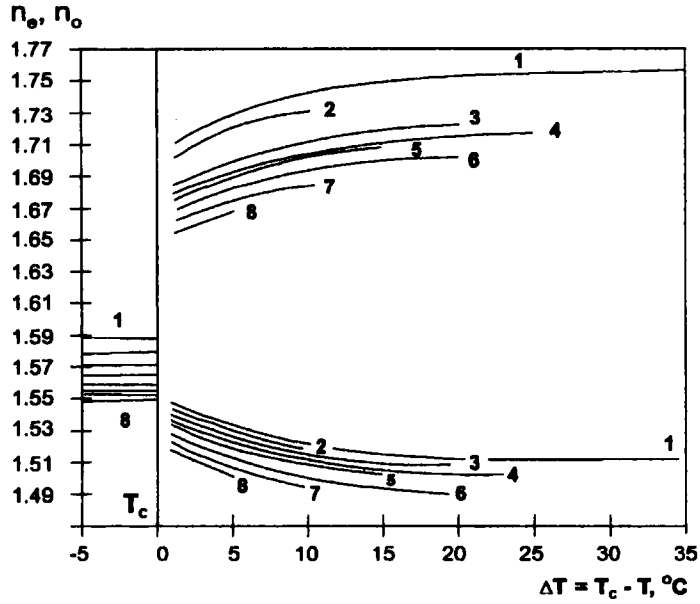


FIGURE 2 Structures, phase transition temperatures and refractive indexes n_e and n_o of eight BAAB homologs

temperature controller gave a temperature stability of 0,01°C. The polarizing microscope Olympus BHMJ was used for observation, the cell was homogeneously illuminated by scattered radiation from Ar^+ laser with help of short length of fiber optic bundle. Orange-colored view of a polarizing microscope after irradiation with Ar^+ laser changed its color to green after one can be observed appearance of the small point-like areas of isotropic phase that were spreading through the view. The dependences of time of photoinduced

Downloaded by [University of Haifa Library] at 20:20 20 August 2012

switching from mesomorphic to isoptropic state τ_{wr} on layer thickness d,

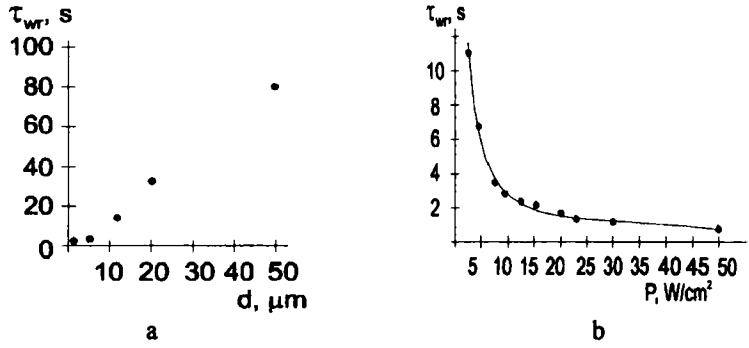


FIGURE 3 Switching times of photochemical nematic-to-isotropic phase transition in BAAB-1 film as a function of its thickness (a) and power of Ar^+ laser (b): a-P=50mW/cm^2, b-d=5μm

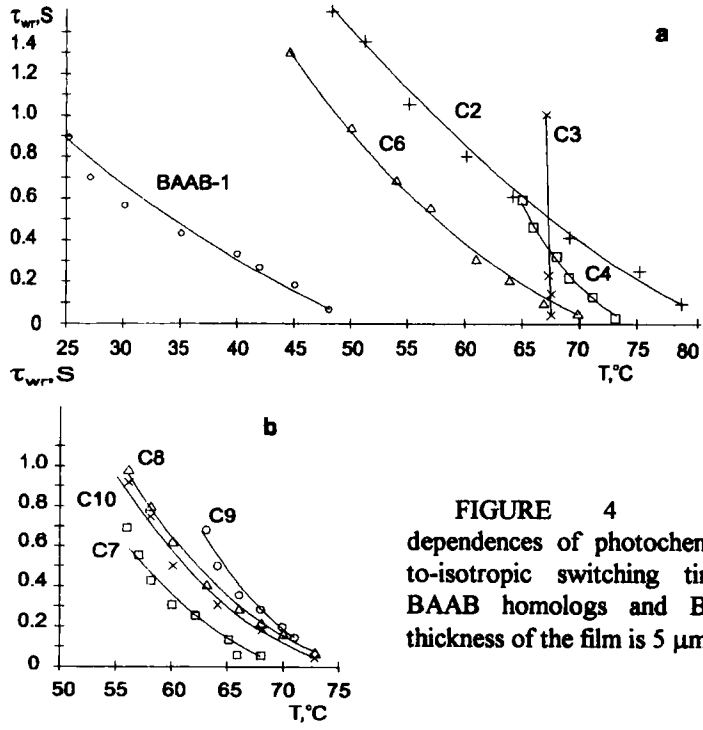


FIGURE 4 Temperature dependences of photochemical nematic-to-isotropic switching time of eight BAAB homologs and BAAB-1. The thickness of the film is 5 μm.

Downloaded by [University of Haifa Library] at 20:20 20 August 2012

excitation power of Ar^+ laser P and temperature T for homologs are show in Fig. 3 and 4. In case of the thinnest samples BAAB-1 with thickness of 0,9μm at the temperature of 23^{0}C the value of τ_{wr} was about 150ms at P=50mW/cm^2. The minimal time of photochemical transition (τ_{wr}=30ms) was observed the temperature thermal phase transition N→I (Δ T=1°C).

On basis of the diffusion equation for non-mesogen molecules N[20]:

$$\frac{dN}{dt} = k_e \alpha_e \frac{P}{h\nu} - \frac{N}{\tau} + D\Delta N$$

one can evaluate their concentration N/N_0, at which the photochemical transition takes place. Here k_e -is absorption coefficient for extraordinary wave ($\mathbf{E}_{Ar} \| \mathbf{L}$), α_e -quantum output of process, P-intensity, τ -characteristic time of cis-trans transition, $N_o = N_A \rho / M$ (ρ is density, M- molecular weight). Because the value of τ for azobenzenes markedly exceeds the switching time τ_{wr} in case of thin cells the second term in equation is negligible and in case of steady distribution of isotropic phase in all of interaction volume we can write $\frac{N}{N_o} = \frac{k_e P t M}{N_A h \nu \rho}$. For the cell filled with BAAB-1 at d=5μm, k_e=300 cm^{-1}, P=50 mW/cm^2, ρ=1g/cm^3, M ≈300 we obtain $N/N_0 \approx 2 \cdot 10^{-2}$. This value of cis-molecules concentration was used for evaluation of dependence τ_{wr}(P) and it was obtained a good agreement with experimental data (the curve in Fig.3).

Azobenzenes, as already it was noted, are characterized by relatively slow relaxation of cis-isomer into initial state. In thick cells the isotropic areas disappeared for a few hour but in thin cells they could exist during a day. For samples having crystalline phase at the room temperature it was observed the freezing of an isotropic area at cooling and the restoring of it after subsequent heating of cell (d=5 μ m). Erasing occurred by existence in darkness or by heating of a cell to temperature above the phase transition point N→I. It was found that quick erasing of isotropic areas can be brought off by focused radiation from He-Ne laser (P=3 W/cm^2).The change of BAAB-1 layer transmission at turning on and turning off of Ar^+ laser is show in Fig. 5. It is

Downloaded by [University of Haifa Library] at 20:20 20 August 2012

seen the full recalling of initial state in 5s. This writing-recalling was good repeated, Fig. 5 demonstrates the three cycles of writing-recalling. In view of polarizing microscope on background of isotropic area 1,5mm across it was observed uniform planar spot that was the size of the He-Ne focused beam diameter (about 200 μ m). The erasing time was increased at heating of layer or at decreasing of beam intensity (Fig.6). The dependence τ_{er}(P) is inversely proportional similarly τ_{wr}(P). At heating of layer τ_{wr} drops whereas τ_{er} increases. Near the phase transition point (Δ T<2 ° C) the radiation power of He-Ne laser was insufficient to return layer into planar state. Reverse process is hindered, that is a result of

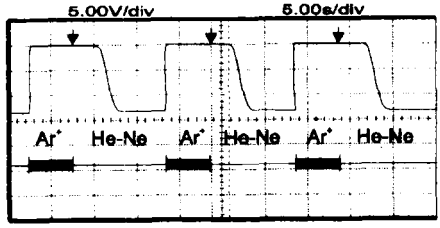


FIGURE 5 Writing (Ar^+ laser) and erasure (He-Ne laser) of optical information on BAAB-1 film based on isothermal phase transition.

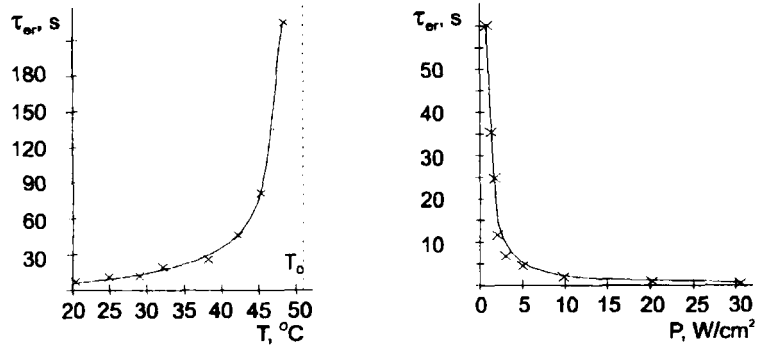


FIGURE 6 Erasure times on BAAB-1 film as a function of temperature and power of He-Ne laser (d=5μm).

abrupt decreasing of order parameter near N→I transition as well as of increasing of thermal diffusion of molecules from surrounding isotropic area inward mesophase area.

Downloaded by [University of Haifa Library] at 20:20 20 August 2012

Downloaded by [University of Haifa Library] at 20:20 20 August 2012

2.Reorientation in the bulk of LC

To research of the orientation phenomena the methods of grating was used. As excitation source it was used the Nd:YAG (FWHM) laser, that operated in second harmonic mode (λ=533nm, W=40mJ/cm^2, Δt=25ns). For cells with thickness of 50μm the medium was transparent at this wavelength, so selfdiffraction can be observed at minor changes of refractive indexes n_o and n_e without inducing of photochemical phase transition. The concentration of cis-isomer was about 10^{-2} mol.%. For experiments we used the cells filled with planar oriented BAAB-1. At λ=533nm and d=50μm the absorption coefficients for e-(L||E) and o-components (L⊥E) were k_e=160 cm^{-1} and k_o=62 cm^{-1}, respectively; the cell was practically opaque to wavelengths of radiation from other lasers (488nm and 440nm). The He-Ne laser (λ=633nm, P=10mW) was used as source of probe beam. The size of spot of interaction was 2mm. At P=80 mW/cm^2 marked influence on reverse cis-trans isomerization was not observed.

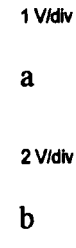


FIGURE 7 Oscilloscope traces of the probe beam diffraction on BAAB-1 sample (d=50μm) induced by Nd:YAG laser. Horizontal scale: a- 1μs/div, b-5μs/div, c-10ms/div.

In Fig.7 the typical oscillograms of change of first-order diffraction intensity are shown. After action of Nd:YAG laser pulse the diffracted beam intensity build-up was observed during 2μs. The relaxation consists of a fast component (25μs) and a slow component (35ms). At the interaction with radiation in azobenzenes one can displayed the thermal, conformational and orientation nolinearities with characteristic relaxation time $\tau = \frac{\rho_o C_p^{\,2} \Lambda^2}{4\pi^2 \lambda_T}$, where C_p is heat capacity, λ_T-

thermal heat conductivity coefficient, Λ - grating period, $\tau_{cf} = \frac{2}{\frac{1}{\tau} + \frac{4\pi^2 D}{\Lambda^2}}$ [20]

and $\tau_\Theta = \frac{\gamma\Lambda^2}{4\pi^2 K}$, where γ -viscosity, K- elastic constant. For the grating with period Λ=11 μm and typical values ρC_p=1.5J/cm^{3}K, λ_T=16J/K·s·cm, K/γ=10^{-6}cm^2/s we obtain τ_T= 28μs and τ_Θ= 30 ms that agree with experiment. τ_{cf} markedly exceeds τ_Θ, its experimental values are presented below.

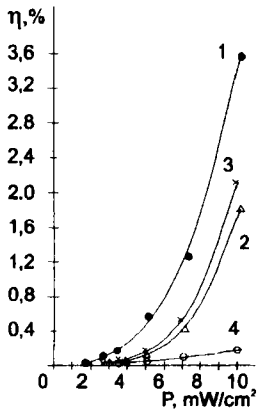


FIGURE 8 Probe beam diffraction efficiency as a function of energy of Nd:YAG laser. The geometries of interection:
1- $\mathbf{L}||\mathbf{E}_{1,2}||\mathbf{E}_{probe}$, 2-$\mathbf{L}||\mathbf{E}_{1,2}$, $\mathbf{L}\perp\mathbf{E}_{probe}$,
3- $\mathbf{L}\perp\mathbf{E}_{1,2}$, $\mathbf{L}||\mathbf{E}_{probe}$, 4-$\mathbf{L}\perp\mathbf{E}_{1,2}$, $\mathbf{L}\perp\mathbf{E}_{probe}$.

We observed the reorientation in BAAB film. The results of investigation of diffraction efficiency η at both various levels of excitation and geometries of interaction are shown in Fig.8. The largest gratings efficiency is reached as the layer director **L** is parallel to polarization vectors of excitation waves $\mathbf{E}_1$, $\mathbf{E}_2$ and probe wave $\mathbf{E}_{probe}$. In accord with $\eta = \left(\frac{\pi\Lambda nd}{\lambda}\right)^2$ the change of refractive index for extraordinary wave $\Delta n_{e||}$ can be evaluated. At the excitation energy 10mJ/cm^2 $\eta_{e||}$=0.036 and $\Delta n_{e||}$=-7.6·10^{-4}. For o-component of probe beam at geometry $\mathbf{L}||\mathbf{E}_{1,2}$ $\eta_{o||}$=0.018 and corresponding value of $\Delta n_{o||}$ is equal to 5.4·10^{-4}. As the layer director is mutually perpendicular to $\mathbf{E}_1$ and $\mathbf{E}_2$, the e-component of probe beam remain high: $\eta_{e\perp}$=0.02, $\Delta n_{e\perp}$=-5.8·10^{-4}, but o-component is minor: $\eta_{o\perp}$=0.002, $\Delta n_{o\perp}$=10^{-4}. In all of cases the diffraction efficiency has the square dependence on pump radiation intensity and is, in general, in accordance with the change of absorption coefficients $k_{o,e}$. As $k_e \approx 2.6k_o$ the value η for e-component must differ from that value for o-component approximately by seven fold if we deal with usual conformational nonlinearity of LC for that $\Delta n_{o,e} \sim k_{o,e}$ [20]. Remind,

Downloaded by [University of Haifa Library] at 20:20 20 August 2012

that in case of conformational nonlinearity the change in $\Delta n_{o,e}$ is due to shift of curves $n_{o,e}(T)$ along the temperature axis. However the observed change of $\eta_{e\|}/\eta_{o\|}\approx 2$, but $\eta_{e\perp}/\eta_{o\perp}\approx 10$ hat is the ratio η_e/η_o do not conserves itself constant as that was observed in works of Odulov et.al.[20] for conformational nonlinearity. The refractive indexes change that is due to reorientation is added to refraction indices change that is due to formation of conformers.

In our case the reorientation occurs in direction perpendicular to initial orientation of layer both in plane of substrates and perpendicular to them when absorption is maximal for e-component of writing radiation. For o-component, as absorption coefficient is less, the rotation is observed mainly in direction perpendicular to substrates. This reorientation is not connected with light field because the Freedericksz-transition-like rotation observed in [21] here is absent otherwise at geometry $\mathbf{L}\perp\mathbf{E}_{1,2}$ for o-component of probe beam the strong signal could be registered. We suppose that reorientation is due to cis-isomers which are formed as a result of inversional mechanism of molecule trans-conformation change [22]. For used materials the conformational and orientational nonlinearities are good distinguished on the basis of relaxation times. At increasing of pump waves power above 10mJ/cm^2 the dependences $\eta(W)$ exhibited the saturation and even decreasing of signal. At this slowly relaxing gratings are observed, that erased themselves in 10 seconds as a result of isomers diffusion. At the same time the area of exposure that was more intensive orange coloured in comparison with basic layer, could exist as long as a few hours. Investigation of those area by the Babinet compensator showed that refractive index of medium is distributed in accordance with Gaussian section of single-mode radiation from Nd:YAG laser, its maximal change caused the shift of an interferention band on a half that corresponds to phase change on π or refractive index change $\Delta n_e = \lambda/2d$ on 0.0063 (P=30mJ/cm^2). At the increasing of excitation energy above 120mJ/cm^2 the photochemical phase transition was observed.

Supposing the time of erasing of isomer grating τ_{cf}=10s in accordance with relationship $\eta \approx \dfrac{\Lambda^2}{\Lambda^2 + 4\pi^2 D_\perp \tau_{cf}}$ one can evaluate the diffusion coefficient for

Downloaded by [University of Haifa Library] at 20:20 20 August 2012

cis-isomers, assuming $\eta \approx 0.03$, $\Lambda = 11\mu m$. Calculated value of $D_\perp$ is equal to $1.5 \cdot 10^{-8} cm^2/s$.

CONCLUSIONS

The writing and the erasure of information on isothermal phase transition from nematic into isotropic phase in 4-n-butyl-4'-n-alkoxyazobenzenes with help of radiation from Ar+ and He-Ne lasers of relatively small power (7mW) was investigated. For practical purposes the mixture BAAB-1 having nematic phase at the room temperature is convenient. For this mixture in the cell thickness of 0.9μm the time of writing was 150ms ($P_{Ar}=50mW/cm^2$) and time of erasure was 1s ($P_{He\text{-}Ne}=3W/cm^2$).

Reorientation of director axis in bulk of BAAB-1 under second harmonic generation of Nd:YAG laser was investigated. We suppose that reorientation has photochemical nature. Under radiation the minor part of molecules (1%) transforms from trans- to cis-isomers during short pulse (25ns). Change of medium refractive index is determined by thermal, conformational and orientational nonlinearities that are good distinguished by their grating relaxation times. For grating with period $\Lambda = 11\mu m$ $\tau_T = 25\mu s$, $\tau_\Theta = 35ms$, $\tau_{cf} = 10s$. Orientational grating efficiency depends on polarization's of acting and probe waves. Molecular reorientation in bulk of LC takes place in directions that are orthogonal to initial alignment both in the substrates plane and the plane perpendicular to them.

Acknowledgments

The authors thank Dr.D.West for technical support.

References

[1.] W.E.Haas, K.F.Nelson, I.E.Adams, *J.Electrochem. Soc.*, **121**, 1667 (1974).

[2.] A.A.Kovalev, G.L.Nekrasov, Yu.V.Razvin, V.A.Grozhik, S.V.Serak, in *Optical Methods of Informations Treatment*, edited by V.A.Pilipovich (Nauka i Technika, Minsk, 1978), p.222.

[3.] K.Ogura, H.Hirabayashi, A.Uejima, K.Nakamura, *Jpn. J. Appl. Phys.*, **21**, 969 (1982).

Downloaded by [University of Haifa Library] at 20:20 20 August 2012

[4.] M.Eich, J.H.Wendorff, B.Reck, H.Ringsdorf, *Makromol. Chem. Rapid Commun.*, **8**, 59 (1987).
[5.] S.Tazuke, S Kurihara, T.Ikeda, *Chem. Lett.*, 911 (1987).
[6.] T.Ikeda, O.Tsutsumi, *Science*, **268**, 1873 (1995).
[7.] J.Sackman, *J.Am. Chem. Soc.*, **99**, 7088 (1971).
[8.] T.Ikeda, T.Sasaki, K.Ichimura, *Nature*, **361**, 428 (1993).
[9.] W.M.Gibbons, P.J.Shannon, Sh-T.Sun, B.J.Swetlin, *Nature*, **351**, 49 (1991).
[10.] K.Jchimura, Y.Suzuki, T.Seki, A.Hosoki, K.Aoki, *Langmuir*, **4**, 1214 (1988).
[11.] Y.Kawanishi, Y.Suzuki, M.Sakuragi, H.Kamezaki, K.Ichimura, *J. Photochem. Photobiol. A : Chem.*, **80**, 433 (1994).
[12.] U.Wiesner, N.Reynolds, C.Boeffel, H.W.Spiess, *Liquid Crystals*, **11**, 251 (1992).
[13.] A.G.-S.Chen, D.J.Brady, *Opt. Lett.*, **17**, 1231, 1992.
[14.] I.C.Khoo, H.Li, Yu.Liang, *IEEE J. of Quant. Electr.*, **29**, 1444 (1993).
[15.] H.Li, Yu.Liang, I.C.Khoo, *Mol. Cryst. Liq. Cryst.*, **251**, 85 (1994).
[16.] T.Marussi, Yu.Reznikov, D.Voloshchenko, V.Reshetnyak, *Mol. Cryst. Liq. Cryst.*, **251**, 209 (1994).
[17.] F.Simoni, O.Francescangeli, Yu.Reznikov, S.Slusarenko, *Opt. Lett.*, **22**, 549 (1997).
[18.] Shulte-Frohlinde D., *Annalen*, **612**, 138 (1958).
[19.] P.V.Adomenas, V.A.Grozhik, *Izv. AN BSSR, Ser. Chem.*, N2, 36 (1977).
[20.] S.G.Odulov, Yu.A.Reznikov, M.S.Soskin, A.I.Khizhnyak, *J. Sov. Phys. JETP*, **82**, 1475 (1982); **85**, 1988 (1983).
[21.] I.Janossy, L.Csillag, A.D.Lloyd, *Phys. Rev.*, **A 44**, 8410 (1991).
[22.] T.Naito, K.Horie, I.Mita, *Macromolecules*, **24**, 2907 (1991).

Downloaded by [University of Haifa Library] at 20:20 20 August 2012