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Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/gmcl20>

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Version of record first published: 31 Aug 2012.

To cite this article: N. V. Kamanina , P. Ya Vasilyev , A. I. Vangonen , V. I. Studeonov , Yu E. Usanov , F. Kajzar & André-Jean Attias (2008): Photophysics of Organic Structures Doped with Nanoobjects: Optical Limiting, Switching and Laser Strength, Molecular Crystals and Liquid Crystals, 485:1, 945-954

To link to this article: <http://dx.doi.org/10.1080/15421400801925851>

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Photophysics of Organic Structures Doped with Nanoobjects: Optical Limiting, Switching and Laser Strength

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Optical limiting of the nanoobjects doped organic electrooptical materials have been studied in the visible and near infrared spectral range. It has been shown that the organic materials with nanoobjects can be used as nonlinear absorbers to protect human eyes and technical devices from high laser irradiation. The systems have been treated at the wavelength of 532, 633, 805, 1047, 1064, 1080, 1315, and 2940 nm. As an additional, the switching parameters of nanoobjects-doped liquid crystal compounds and improvement in the laser strength of the materials used as substrates for limiters and switchers have been shown.

Keywords: fullerene; laser strength; liquid crystal; nanotubes; nonlinear transmission; optical limiting; organic electrooptical structures; switching

The authors wish to thank Dr. A. P. Zhevlakov (Vavilov State Optical Institute, St. Petersburg, Russia), Dr Yu. M. Voronin, and Dr. S.E. Putilin (State Institute of Fine Mechanics and Optics, St. Petersburg, Russia). This work was partially supported by ISTC Project IPP A-1484.

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1. INTRODUCTION

At present time the great interest has been expressed in photophysics properties of nanoobjects, such as fullerene, carbon nanotubes (CNT), nanoparticles, J-aggregates, nanofibers, etc. and of organic conjugated systems doped with them [1–3]. The promising nonlinear optical [4–6], photoconductive [7–9], and laser-induced dynamic properties [10–12] can be activated in the organic materials, as well as the self-organization phenomena [13] and increase in the polarizability [14,15] and order parameters [16] can be found under condition of nanoobjects sensitization. Some evidence to form the quasi-photonic organic systems have been shown and presented in the papers [17,18].

As the results of these facts, the wide area of optoelectronics application should be found when the organic systems have been doped with nanoobjects and than these systems have been studied at different wavelength in the visible and IR spectral range, for example, at the wavelength of 532, 633, 805, 1047, 1064, 1080, 1315, 1500, 2940 nm. Thus, the organic materials with nanoobjects can be used as nonlinear absorbers to protect human eyes and technical devices from high laser irradiation; these systems can be considered as effective switchers, converters and modulators of laser beam. Moreover, these structures are useful to apply them in medicine and display technique.

In the present paper the systems of 2-cyclooctylamine-5-nitropyridane (COANP), 2-(*n*-prolinol)-5-nitropyridine, polyimide, polyaniline, polyvinyl alcohol (PVA), polymer-dispersed liquid crystals (PDLC) based on the structures mentioned above have been studied as effective optical limiters, and high speed switcher of the laser beam in the visible and in the IR spectral range. Moreover, some evidences of the laser strength improvement of the intermediate layers used in the limiters and switchers have been shown.

2. EXPERIMENTAL CONDITIONS

The laser experiments were performed with 3–6.10% solutions of nanoobjects-doped electrooptical components in tetrachloroethane, chloroform, benzene, and water. The organic photosensitive matrixes were sensitized by fullerenes (C_{60} and/or C_{70}) with content from 0.1 to 5 wt %, and by CNTs with content from 0.1 to 1.0 wt %. Thin films had thicknesses of 1–5 μm , the solutions or gels were studied in optical chamber with a thickness varied from 10 to 30 mm, and an electrooptical nematic liquid crystal (NLC) layer from cyanobiphenyl group with positive values of the optical and dielectric anisotropy had the thickness of 10–12 μm .

The CW He-Ne-laser, pulsed nanosecond neodymium laser, pulsed nanosecond Er^{3+} :YAG-laser, pulsed nanosecond photodissociative iodine laser, femtosecond quasi-CW Ti-sapphire laser were used to study optical limiting (OL). The experiments were performed at wavelength of 532, 633, 805, 1047, 1064, 1080, 1315, and 2940 nm. Beam energies incident on and transmitted through the sample were measured. The input energy density in the OL experiments was varied in the range of $0.05\text{--}5 \text{ J}\cdot\text{cm}^{-2}$. To study the high speed switching of nanoobjects-doped PDLC structures the experiments were performed at 633 nm with LC cells of the S-type with an initial planar orientation. The drops of the photosensitive component dispersed in the NLC matrix had diameters within 10–100 nm. Orientation of the NLC layer was provided by different method, for example, by rubbing technique, by holographic grating formation, by treatment of surface electromagnetic wave [19]. Last way leads to decrease the applied voltage retaining switching parameters. The switching characteristics were studied by measuring the changes in reaction and relaxation time of the electrooptical response. It has been registered at the first transmission oscillation and detected a transition to the neighboring extremum of the *S*-curve, which corresponded to a change in the phase shift by π . The cell has been treated by rectangular pulses with an amplitude of $A = 10\text{--}30 \text{ V}$, a pulse duration of $\tau_{\text{sup}} = 1\text{--}10 \text{ ms}$, and a repetition frequency rate of $1/T = 0.5\text{--}200 \text{ Hz}$.

3. RESULTS AND DISCUSSION

The basic results of the OL study in the visible and near infrared range are placed between the investigations of other scientific groups and are shown in Table 1. Different mechanisms, such as reverse saturable absorption, light-induced charge-transfer complex (CTC) formation, plasmon resonance, laser-induced heating, laser-induced scattering, multiphoton absorption, carriers free absorption, and others are considered as the main factors determining the OL in the visible and in the near-IR spectral ranges [20–34]. As an additional effect that influences the laser attenuation due to energy losses via diffraction has been taken into account too. Really, it has been established in papers [6,14,21,35–37] for the materials doped with nanoobjects that the change in the laser-induced refractive index has been corresponded to 10^{-3} and more. This value is two orders of magnitude larger than that observed due to thermal nonlinearity. Thus, the photorefractive grating formation should be considered as the OL mechanism too. It should be noticed that for IR OL the CTC formation

TABLE 1 Comparative Data on Visible and Infrared Optical Limiting

Systems	Initial transmission, %	Wavelength, nm	Pulse duration, ns	OL threshold, J · cm ⁻²	Destruction level J · cm ⁻²	Ref.
Composites with Ag nanoparticles		3800–4200	250	0.005–0.025		[20]
2-(<i>n</i> -prolinol)-5-nitropyridine-C ₆₀	65–70	2940	500 μs	0.9–1	≥1.5	[12]
Polyimide-C ₇₀	~80	1315	50	0.6–0.7	~1.5–2	[21]
Polyimide-C ₇₀	~80	1315	50	0.08–0.1 (with keep in mind the Xe-lamp irradiation, which pump iodine laser)	~2	[22]
Polyimide-C ₆₀	70–75	1080	10	0.7	~2	Present
Polyimide-C ₇₀	75	1080	10	0.75–0.8	≥2.5	Present
Polyimide-CNTs	75–80	1080	10	0.05–0.0.1	3	Present
Liquid crystal-polyaniline-C ₆₀	70	1080	10	0.1–0.3	≥0.5	[23]
Stabilized gels based on PVA-CNTs	10–70	1080	10	0.07	>5	[24]
CNTs solution in tetrachloroethane	50–70	1080	10	0.9–2	>5	[24]
Suspension of carbon nanoparticles in water and CS ₂	~80	1064	10	0.12–0.7		[25]
Zn-phthalocyanine-C ₆₀	75–80	1064	Some ns			[26]
Suspension of carbon nanotubes in water and chloroform	≥90	1064	6	0.15–0.35		[27]
Polyimide-C ₇₀	~79–85	1047	8	0.6–0.7	~2.5–3	[28]
Mg-phthalocyanine-C ₆₀	70	1047	8	1.5	>3	[29]
Liquid crystal-COANP-C ₇₀	65	805	70 fs		>0.5	[30]
COANP-C ₇₀	65–67	805	50 fs		>1	Present
Liquid crystal-polyaniline-C ₆₀	67.5–89	805	30 fs	0.09–0.25	>0.5	[24]
Solution of C ₆₀	84	710–740	10	2		[31]
C ₆₀ -SiO ₂ -sol-gel	75	532	30	0.3	4	[32]
Solution of C ₆₀	66	532	30	0.1–0.5		[33]
Polyimide-C ₇₀	70	532	10	0.3–0.5	≥2.5	Present
COANP-C ₇₀	70–75	532	10	0.2–0.35	≥2.5	Present
<i>N</i> -(4-nitrophenyl)-(L)-prolinol-C ₆₀	75–80	532	15	0.9–1.5	≥2.5	[34]
<i>N</i> -(4-nitrophenyl)-(L)-prolinol-C ₆₀		337	10	0.35–0.4	≥1.5	[34]

has been indicated as the dominant OL mechanism. To support CTC formation different evidences can be done. For example, for the systems studied in the present paper these evidences have been found and shown in the publications [38–40].

It should be mentioned from our previous results that the large value of the laser-induced change of the refractive index can be estimated and the nonlinear refraction n_2 and third order susceptibility $\chi^{(3)}$ have been found. Nonlinear refraction and third-order susceptibility are: $n_2 \sim 10^{-10} \text{ cm}^2 \text{ W}^{-1}$ and $\chi^{(3)} \sim 10^{-9} \text{ esu}$ for the thin-film of nanoobjects-doped structures and $n_2 \sim 10^{-9} \text{ cm}^2 \text{ W}^{-1}$ and $\chi^{(3)} \sim 10^{-8} \text{ esu}$ for nanoobjects-doped PDLC. The value of $\chi^{(3)}$ is responsible for the change in the local volume polarizability and predicts the drastic improvement in the switching characteristics of the nanoobjects-doped PDLC. Really, using experimentally observed increase in the diffraction efficiency, both n_2 and the nonlinear susceptibility $\chi^{(3)}$ could be calculated from the following equations [41]:

$$n_2 = \frac{\Delta n_i}{I}, \quad (1)$$

$$\chi^{(3)} = \frac{n_2 n_0 c}{16\pi^2}, \quad (2)$$

where n_0 and n_2 are linear and nonlinear refractive indices, respectively; Δn_i is the laser-induced change of the refractive index; c is the velocity of light; and I is the intensity of the light wave. Thus, for example, n_2 and $\chi^{(3)}$ for PDLC based on 5 wt.% C₇₀-doped COANP are $1.6 \times 10^{-9} \text{ cm}^2 \text{ W}^{-1}$ and $4.86 \times 10^{-8} \text{ esu}$, respectively, at the incident energy density of $17.5 \text{ mJ} \cdot \text{cm}^{-2}$ and $\Delta n = 1.4 \times 10^{-3}$. This data observed when the systems studied under four-wave mixing technique at wavelength of 532 nm. The data obtained are close to those for silicon (10^{-10} and 10^{-8} , respectively). It should be noticed that the nonlinear effect in the structures under present investigation could be accumulated and hence increased significantly. It should be noticed that these values are significantly larger estimated for pure or dye-doped structures. It should be noticed that the increase in $\chi^{(3)}$ determines the increase in polarizability of the nanoobjects doped systems, thus it predicts easy control of high speed switching of them. Really, let us consider a small local volume with dimensions less than the incident irradiation wavelength. For our system the wavelength is 532, 633, 1064, etc. nm and the nanoobjects dimensions, for example, fullerenes, are 0.65–0.7 nm. In this case the induced dipole is the most important optical characteristic. Its dependence on the local field

applied is expressed in terms of the dipole polarizability $\alpha^{(n)}$, which is directly proportional to the nonlinear susceptibility $\chi^{(n)}$ and to the unit cell volume ν [42]:

$$\chi^{(n)} = \alpha^{(n)} / \nu \quad (3)$$

As it has been noticed before this value is significantly larger than that estimated for pure or dye-doped structures. For example, for recording a thin amplitude-phase hologram at wavelength of 532 nm on the PDLC based on COANP-fullerene, the response in the first diffraction order changes from 2×10^{-4} to 3.7×10^{-3} mJ as the incident energy density of the nanosecond pulsed laser irradiation changes from 2.5 to 17.5 mJ \cdot cm $^{-2}$. The response in non-sensitized LC is at least 2 orders of magnitude less. Therefore, using the fact that polarizability of all structures can be accumulated from local volumes, it can be found that:

$$P = \chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \dots + \chi^{(n)}E^n + \dots \quad (4)$$

The high value of nonlinear coefficient $\chi^{(3)}$ demonstrates potentials to control the switching parameters of the PDLC devices with good advantage. The introduction of nanoobjects, for example, an organic (donor) – fullerene (acceptor) complex, representing a charge-transfer complex with an additional dipole moment and increased local polarizability (per unit volume of the sensitized medium), can accelerate the process of reorientation (switching) of molecules in the system. The creation of this additional dipole moment is probably favored

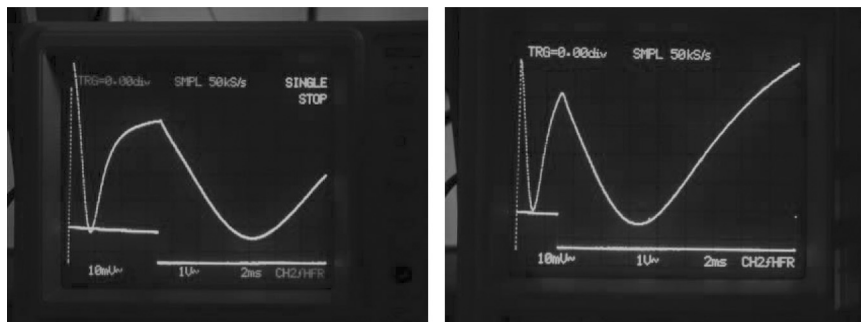


FIGURE 1 Electrooptical response of nanoobjects-doped PDLC based on COANP-fullerene structure at the parameters of pulsed supply voltage: amplitude is 12 V (left figure) and 17 V (right figure); repetition frequency is 50 Hz; pulse duration is 3 and 2 ms, respectively.

TABLE 2 Improvement of the Laser Strength Properties of ITO Contacts in the LC Switching Element due to Nanotubes Placing and SEW Treatment

Type of the layers	Laser energy density, $J \times cm^{-2}$	Laser energy density destroyed layer, $J \times cm^{-2}$	Number of pulse before destroy, $J \times cm^{-2}$	Remarks
Pure ITO	0.35–0.5	0.65–0.67	10 at 0.66	At this laser energy density the output signal has been changed on 50%.
ITO with SEW	1.025–1.05	1.25	7–10 at 1.25	
ITO covered with HfO_2	0.375–0.6	0.67–0.7	10 at 0.7	
ITO covered with HfO_2 than treated with SEW	0.88–1.06	1.18–1.25	7–10 at 1.2	
ITO covered with nanotubes	0.4–0.7	0.75	10 at 0.75	At this laser energy density the output signal has been changed on 10%.
ITO covered with nanotubes than treated with SEW	0.94–1.25	1.5–1.56	10 at 1.5	

by the effective charge generation in the structures under the action of light, which enhances the effective charge separation between the donor part of organic molecules and nanoobjects as acceptor, thus increasing the polarization of the whole system. Thus, the use of nanoobjects, for example fullerenes, ensures the transition from the milli- to microsecond range of switching parameters, which is characterized the smectic LCs. It should be mentioned that the commercial NLCs used by different LC companies to develop new display elements reveal the switching parameters in the range of 4–16 ms at the thickness of the element of 1–3 micrometers. The introduction of the nanoobjects in the nematic LC stimulates the decrease in the time-on and time-off characteristics up to 0.5–1 ms. The Figure 1 shown these results, which predict that nanoobjects-doped PDLCS can be considered as display elements of new generation.

It should be mentioned, that when we have treated the nanoobjects-doped PDLC, the increase in the laser strength has been found when interface between LC mesophase an solid substrate has been modified with nanotubes. The data are shown in the Table 2.

Due to larger number of C–C bond in the nanoobjects we have observed the high laser strength of the ITO contacts with nanotubes and studied the laser strength of the ITO layers with and without nanoobjects placing. Analyzing the Table 2 results, one can see that the laser strength of the ITO covered with nanotubes and than treated with surface electromagnetic wave (SEW) revealed the best laser strength. It should be mentioned, that this treatment has been made when glass or quartz substrates have been used. In this case at the level of energy density close to $1.5 \text{ J} \times \text{cm}^{-2}$ the output signal has been changed not more than on 10%. The SEW source was a quasi-CW gap CO_2 laser generating *p*-polarized radiation with a wavelength of 10.6 micrometers and a power of 30 W. The skin layer thickness was ~ 0.05 micrometers.

4. CONCLUSION

Firstly, we have studied the influence of nanoobjects-doped systems on the optical limiting properties of the organic materials at different wavelength in the visible and near-infrared spectral range. Secondly, we have observed high speed switching in the nanoobjects-doped LC, when the traditional nematic LC have been used with classical switching characteristics for this nematic from cyanobiphenyl group close to 4–16 ms. Nanoobjects doping increases the speed of LC elements up to 0.5–1.5 ms. The effect is correlated with increase in the polarizability of the nanoobjects-doped compounds. Thirdly, we have improve the

laser strange of the interface between solid and LC that permits to use the modified structures in the laser techniques with high energy density with good advantage.

The direction of research is of interest both from the standpoint of fundamental and of practical applications of novel nanomaterials and due to the possibility to apply them as real optoelectronic elements.

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