JOURNAL OF OPTOELECTRONICS AND ADVANCED MATERIALS Vol. 8, No. 1, February 2006, p. 172 - 176

The effect of neutron irradiation on fatty acids and computer modelling

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The goal of this study is to model the changes induced by thermal neutron irradiation in some fatty acids with important biological role. These changes could be experimentally evidenced by the nonlinear optical answer of the samples under non-destructive laser light, while in the liquid crystal state, a state involved in many mechanisms from the living matter. The experimental data were processed with Table Curve3D computer program that gives the possibility to forecast the behavior of other samples and to model it with different equations. For simulating the dynamics of the phenomena inside the material, a model based on Runge-Kutta functions in MATLAB was used. This method is adequate for modeling the interaction of different external signals with different materials, by using appropriate parameters and time intervals.

(Received October 14, 2005; accepted January 26, 2006)

Keywords: Fatty acids, Liquid crystals, Dielectrics, Nonlinear optical response, Thermal Neutron irradiation, Computer modeling

1. Introduction

The mesomorphic liquid crystals (LC) found many applications in the fundamental research, in technology, biology, medicine, pharmacy etc. Continuation and extension of the study of mesomorphic LC is of high importance for the living matter, since it seems that LC state is involved in the response to stimuli from the environment [1].

Thin films of saturated and unsaturated fatty acids (FA), components or forerunners of the biological membrane (BM), in mixture with cholesterol (Ch) or some drugs (like aspirin) were investigated as simple BM models, while in the LC state: butyric acid (4:0), caproic acid (6:0), caprylic acid (8:0), capric (10:0), lauric acid (12:0), myristic acid (14:0). stearic acid (18:0), arahidic acid (20:0), elaidic acid (18:1), linolenic acid (18:3); arahidonic acid (20:4), (5,8,11,14). The samples were prepared in our laboratory at "Politehnica"University and exhibited different LC textures during their evolution between some characteristic temperature values. The electric conduction and molecular arrangement (textures) of these systems depend on their chemical composition and present memory effects and other modifications under some stimuli from the environment.

Experimental data were processed by computer, using Table Curve3D [2] in order to fit the data and forecast some other experimental situations. Runge-Kutta equations in Matlab were used [3] for simulations and characterization the external incident and emergent signals from the samples (fluctuations, noise, vibrations etc).

2. Preliminary considerations and experimental results

The results obtained by laser radiation (λ = 6328 Å) scattering in these systems [4] led, on the basis of the theory developed by Stein et al. [5] for polymers and extended by M. Honciuc [6] to LC, to the conclusion that our samples are uniaxial with the optic axis oriented on the direction of the long molecular axis. Last years, a special interest in laser applications for investigating different systems belonging to the living matter and for treating many diseases was observed [7 - 9].

Study of FA and mixtures behavior in electric and magnetic field allows for obtaining information on the molecular arrangement, the electric anisotropy, electric membrane potential, magnetic susceptibility, ordering effects.

Experimental results concerning the microscopic aspect of FA and mixtures with Ch or some drug samples, in integral incoherent and in laser coherent light beams, and under some micro particles beams are presented. Some nonlinear optical effects occurring even at low continuous wave laser power or in pulsed laser beams, were emphasized, depending on the: saturated/unsaturated character of the acid, of the impurities amount, and of external perturbation. Typical sandwich LC cells were built and some samples were exposed to a fluence of thermal neutrons irradiation (TNI). As a result of TNI, an essential change in texture and in the electric state of the samples was observed. The electric properties of such systems essentially depend on changing the amount of Ch

in the system. At higher than 11.2 mW laser power, a change of the emergent beam feature was observed for the non-irradiated (NI) samples, similar with the ones observed by us for some irradiated (IR) mixtures of FA–Ch. When the laser power overcomes 12 mW, a ring pattern due to the self-phase modulation of the beam was observed in far field for the NI, but not for the IR samples, even after many laser exposures. After a second irradiation at the end of this interval, the transmission was almost the same as after the first laser light impact.

The hysteretic curves of the current versus the applied voltage exhibited a nonlinear dielectric feature and a negative resistance, displaced to smaller voltage values when Ch was added. This behavior is similar to the one encountered in dielectrics exhibiting a spontaneous polarization [10] and is illustrated in Fig. 1, 2, which allows for the determination of the relaxation time of charge carriers inside the material, the space charge within the sample, and the internal resistance [11 - 13]. All the measurements took into consideration the Ch percentage and the discussion of the results has been done with respect to the hypothesis that Ch acts similarly at an external electric field, applied on the sample.

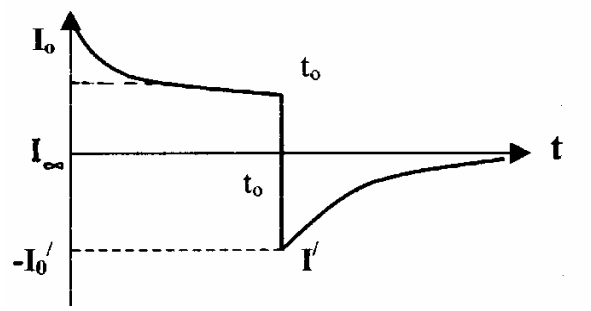

Fig. 1. Theoretical dependence I=I (t) at U=const. and U=0.

The dependence $I = I(t)$ obtained based on the relation:

$$j(t) = j_0 \exp(-t/\tau) + j_\infty \qquad (1)$$

The experimental curves are in a good agreement with the theoretical model.

It is known that all the mesogenic compounds possess a high optical nonlinearity that can be pointed out even at low optical laser powers [14]

The thyeoretical basis of the relation between the electrical and optical measurements is the modification of the dielectric constant and of the refractive index with the composition of the material.

The large change of the refractive index with the light intensity leads to the possibility of the self-focusing of the laser beam and other effects - such as the optical activity change with the intensity of the light and the change of the laser pulse width after passing through the sample. The nonlinear optical effects are shortly reviewed

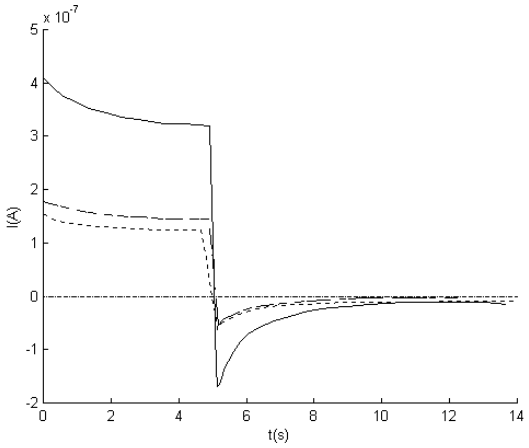

Fig. 2. Dependency I=I (t) for U=5V, Δt=5s, θ=20° C for samples A (dashed line), B (dotted line) and C (continuous line). A - mixture of arachidic—lauric—butyric acids in molar percentages: (0.50÷0.25÷0.25)), sample B - mixture of arachidic—lauric—butyric acids and cholesterol in molar percentages (0.45÷0.25÷0.15÷0.15)); sample C - mixture of arachidic — lauric — butyric acids and cholesterol in molar percentages (0.25÷0.25÷0.25÷0.25)).

The self-focusing has been obtained in UFA cells, and a typical ring pattern in far field has been noticed by using a He-Ne laser (running at 6328 Å and 20 mW).

The pulse width modification in time domain has been emphasized and presented in Fig. 3 by using the TC3D program. A Nd^{3+} glass laser, and a KDP crystal have been used for obtaining a visible light with 523 nm, and the output feature was displayed on a sampling oscilloscope. One can see the effect of the Ch amount in mixtures of UFA, which can develop a method to estimate this amount by means of the output pulse width [11, 15].

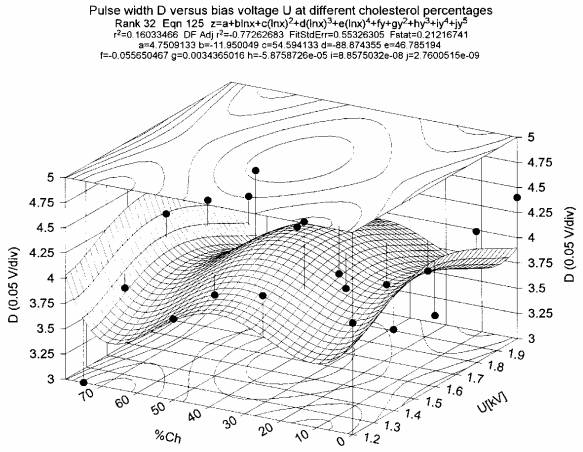

Fig. 3. Pulse width D versus bias voltage U at different cholesterol percentage. Nd^{3+} glass laser SOLARS S-7, IFTAR Bucharest, TEM_{002}, 200 μs, 1.06 μm (SHG 532 nm); sampling oscilloscope TEKTRONIX 7613 (0.05V/div. and 0.5 ms/div.).

Microscopic measurements were performed, too. Each sample has been placed under a microscope that allows visualization of micro structural images in polarized light. Samples are smectic LC, i.e. their molecules have the tendency to form layers, where they are parallel, normally or tilted with respect with the layers' plan. The smectic mesophase shows different states: smectic A, smectic C, SC*, H, G, M etc., depending on the temperature values, and on the electric applied field at constant temperature. The polymorphism of the smectic phase can be explained on the basis of the molecular distribution in the smectic plane. This distribution determines some modifications connected with the smectic layer width and with the distance between consecutive layers.

For T = ct. a certain extern stimulus, like for example the electrical field of the order of 10^4-10^5 V/m or a neutrons beam of 4.15×10^{12} neutrons/cm^2 gives rise to a so called memory effect. In Fig. 4 are presented the corresponding textures for the arachidic acid, under the influence of the electric field. The memory effect, for the same fatty acid, under the influence of a TNI is presented in Fig. 5. One observes the existence of some profound analogies regarding the behaviour of the LC under different external stimuli. This fact suggests the existence of a unitary explanation upon the transition mechanism from one state to another. Considering a microscopic scale, such transitions takes place because a co-operative interaction among different parts of the system, which under some specific conditions can lead too abrupt changes.

Fig. 6 presents the emergent laser beam power versus the incident one and C carbon atom number in non-irradiated (N) pure acid samples (a), irradiated (I) samples (b) at P_{max}=12 mW, λ=6328 Å. By analyzing the graphs, one can state the following: almost all the samples present a nonlinear optical dependence of the output versus input laser power, and an optical hysteresis. Fig. 6 evidences the changes occurred in the sample I, especially for a large C number.

Taking into account these results and previous ones for other saturated and unsaturated FA and mixtures, we believe that TNI led to changes in the molecular arrangement [16, 17], a shift of the molecules from the smectic plane, and modification of the electric state of the samples. The presence of Ch modifies this process. The length of the molecule and the distance between the double bound and the carboxyl group is also important. Relatively low undestructive laser power is useful for emphasizing the changes of the structure and properties in TNI samples, by their nonlinear interaction with these substances, easy to obtain in the LC state.

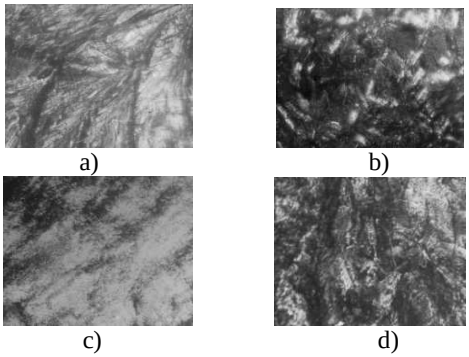

Fig. 4. Memory effect for the arachidic acid under the influence of the electric field: a) without electric field; b) after field application; c) after field removal; d) after the second application of the electric field.

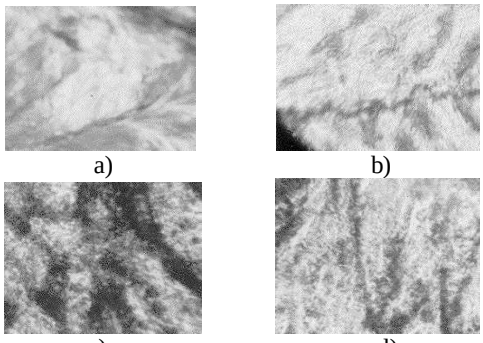

Fig. 5. Memory effect for the arachidic acid under the influence of thermal neutron irradiation. a), b), c), d) illustrate the same steps as above, but in neutron field.

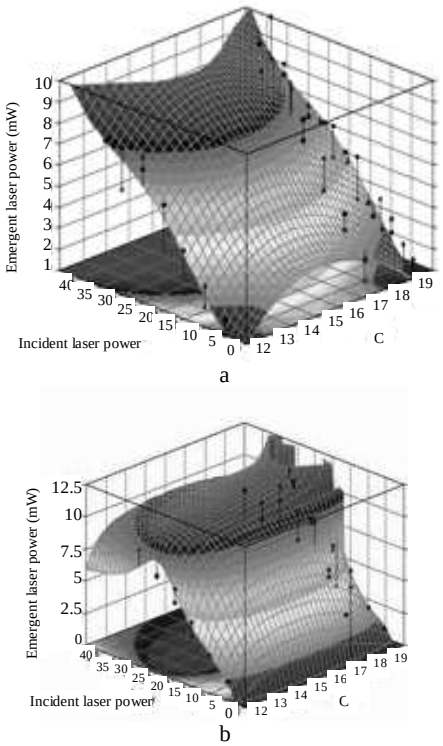

Fig. 6. Output laser power versus input laser power and carbon atom number; a)-NI samples; b)-IR samples.

3. Computer study of the experimental results

The optical output power versus the input one and Ch percentage in mixtures was analyzed with the TC3D software program, by using Surface-Fit-All-Equations option. Then the Numeric Summary option displayed all numerical information, including goodness of fit criteria, coefficient standard errors and confidence limits for the fitted parameters, function extreme, the fitting method, an analysis of variance, and data table statistics. Precision Summary option can be used to determine how much precision is preserved in the current equation. The residual Graph is a separate window displaying the residuals for the current surface-fit. The data table can be weighted, if necessary. Thus, the program gives the possibility to choose other experimental conditions in terms of the future experiment requirements and purposes. The dependencies are generally hysteretic showing a memory effect of the samples.

We were looking for a mathematical model for describing the dynamics of the phenomena taking place inside the material, under the influence of external optical pulses. Due to the high optical frequency, we cannot use a linear equation of evolution, which response would be close to zero. We are looking for a nonlinear equation of evolution, able to generate pulses similar to test-functions. These functions, similar to a Dirac pulse, can be written under the form:

$$\varphi = exp\ [a^2/(\tau^2 - 1)] \qquad (2)$$

where $\tau = t - t_m$, *and* t_m is the middle of the working period. Such a function has nonzero values only for $\tau \in [-1, 1]$. We are looking for a differential equation, which can have as a solution the function φ. However, such an equation cannot generate the test function φ; the existence of such an equation of evolution, beginning to act at an initial moment of time, would involve the necessity for a derivative of certain order n - noted $f^{(n)}$ to make a jump between two null values. This is in contradiction with the property of the test-functions to have continuous derivatives of any order on the whole real axis, represented by the time axis. In this case, we try as a solution of this equation a function *f* with nonzero values on the interval $\tau \in [-1, 1]$, and a certain number of continuous derivatives on the whole time axis. Therefore, we study those evolutions depending only on the values *f*, $f^{(1)}$,…, $f^{(n)}$, equal to the values of φ, $\varphi^{(1)}$, ..$\varphi^{(n)}$ at a certain time moment, very close to the initial moment $\tau = -1$.

One can write the equations:

$$f^{(1)} = [-2\tau/(\tau^2 - 1)]\ f \qquad (3)$$

$$f^{(2)} = [(6\tau^4 - 2)/(\tau^2 - 1)] \qquad (4)$$

$$f^{(2)}(\tau) = [(0.6\tau^4 - 0.36\tau^2 - 0.2)/(\tau^2 - 1)^4]\ f(\tau) \qquad (5)$$

With different initial conditions for *f*, $f^{(1)}$ and using Runge-Kutta functions in MATLAB, it resulted that the equation that leads to some functions similar to a rectangular unitary pulse is the last one, since the amplitude is close to unity for more than 2/3 of the integration period.

However, in our case, all changes inside the material appear due to the external optical pulse. Therefore, we must consider null initial conditions for the system, and add a "free" term in the differential equation – corresponding to the magnitude of the electrical field of the external signal, having a frequency of about 10^{15} Hz. The working period was chosen approximately equal to the period when the optical signal is received by the detector (about 0.2 ms). The differential equation can be written as:

$$f^{(2)} = [(0.6\tau^4 - 0.36\tau^2 - 0.2)/(\tau^2 - 1)^4]\ f + u(t) \qquad (6)$$

where u is represented by an alternating function with a frequency 10^{11} times greater than the working period of 0.2 ms. By numerical simulations in MATLAB with Runge-Kutta functions, we have obtained for f the results presented in Fig. 7 for $u = cos(10^{11}\pi\tau)$.

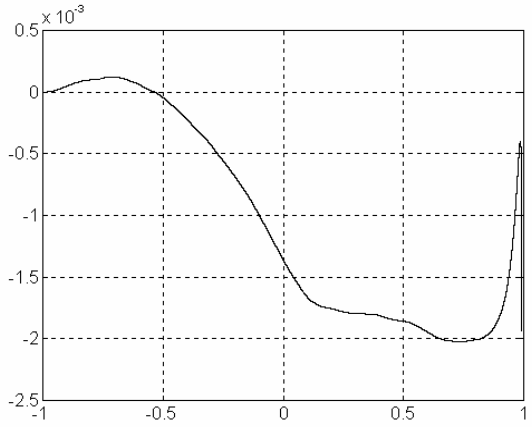

Fig. 7. f versus t for $u = cos(10^{11}\pi\tau)$.

The function *f*, generated by the material under the influence of the external optical pulse, can be integrated on this working time, the result of this operation representing the physical quantity measured by the external observer.

It can be noticed that, for the external observer, the behavior of the material presents a slowly varying evolution in time-a single oscillation on the whole working interval, even though the input signal is a fast varying one (at an optical frequency), similar to the behavior noticed during the experiments. This allows to consider our method as appropriate, if based on the use of systems described by differential equations, to generate pulses similar to test functions. In fact this is a general method that could be applied in different situations of interaction of some signals with material nonlinear media, by an appropriate choice of the parameters and working period.

The experimental observations are in accordance with some models and simple theoretical assumption existent in the literature [19 - 21] or developed by us.

4. Conclusions

We analyzed some systems with important biological role. The study underlined the influence of the chemical composition (carbon atom number, saturation feature, Ch content) of the systems from the living matter, and the possibility to connect this information with the electric or nonlinear optical behavior of these systems. External perturbation from the environment changed the state of these systems, evidenced by using the same connection between the structure and the electric and nonlinear optical behavior of the samples.

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