Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

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P. R. G. Fernandes[a]; H. Mukai[a]; B. S. L. Honda[b]; S. M. Shibli[b]
[a] Departamento de Física, Universidade Estadual de Maringá, Av. Colombo, 5790, 87020-900 - Maringá, Paraná, Brazil [b] Instituto de Física, Universidade de São Paulo, Caixa Postal 66318, 05389-970 - São Paulo, Brazil

To cite this Article Fernandes, P. R. G. , Mukai, H. , Honda, B. S. L. and Shibli, S. M.(2006) 'Induction of order in the isotropic phase of a lyotropic liquid crystal by pulsed light', Liquid Crystals, 33: 3, 367 — 371
To link to this Article: DOI: 10.1080/02678290600583227
URL: http://dx.doi.org/10.1080/02678290600583227

Liquid Crystals, Vol. 33, No. 3, March 2006, 367–371

Induction of order in the isotropic phase of a lyotropic liquid crystal by pulsed light

P.R.G. FERNANDES*†, H. MUKAI†, B.S.L. HONDA‡ and S.M. SHIBLI‡

†Departamento de Física, Universidade Estadual de Maringá, Av. Colombo, 5790, 87020-900 – Maringá, Paraná, Brazil

‡Instituto de Física, Universidade de São Paulo, Caixa Postal 66318, 05389-970 – São Paulo, Brazil

(*Received 15 November 2004; in final form 3 February 2005; accepted 12 December 2005*)

A surprising observation of order induced in the disordered (isotropic) phase of the lyotropic KL/DeOH/water mixture was experimentally verified using a pulsed laser beam. This effect is reported here for the first time, to the best of our knowledge. We present a theoretical approach based on a diffusion-like equation that is in good agreement with the experimental results.

1. Introduction

Lyotropic liquid crystals are obtained when some amphiphilic molecules are mixed in a solvent, usually water. Micelles are formed when the amphiphilic molecules achieve a critical micellar concentration. The isotropic phase of lyotropic liquid crystals does not have long range orientational ordering, but it is possible to induce birefringence by flow. Some efforts have been made to understand the behaviour of complex fluids under flow [1, 2] and micellar complex fluids are only beginning to be investigated [3–5]. Flow-induced order has been seen in the isotropic phase of thermotropic liquid crystals [6] and has recently been studied in lyotropic systems [7, 8].

The phase diagram for mixtures composed of potassium laurate (KL), decanol (DeOH) and water shows a nematic region surrounded by isotropic domains [9]. In general, these mixtures have the capability to react to the slightest external stimulus, and have a tendency for restoration of the initial order after its removal [10]. The isotropic phase (I) of our lyotropic mixture (KL/DeOH/water) is located between 10 and 40°C, surrounded by two lamellar phases and a nematic region. We verified the existence of birefringence induced by flow in the I phases of this mixture. This effect has been explained using an order diffusion approach [7], and a characteristic length ($\sim 10^{-5}$ cm) was obtained.

From a technological point of view, the induced order effect in I phases can be applied in mechanical vibration sensors based on lyotropic liquid crystals [11]. In this work we report the effect of the induction of order by pulsed light in the isotropic phase of the lyotropic liquid crystal. In § 2 this effect is explained using a theoretical approach based on a diffusion-like equation. Experimental results are presented in § 3 and discussed in § 4; conclusions are given in § 5.

2. Theory

The order parameter is a fundamental quantity in the study of phase transitions [12]. In general, this quantity vanishes on one side of the transition and moves away from zero on the other side. In liquid crystals the isotropic phase is characterized with zero order and the nematic phase with an order different from zero. The Landau–de Gennes theory characterizes the equilibrium state of the macroscopic nematic system using a tensorial order parameter Q. In [7] the induced scalar order parameter in the isotropic phase satisfies a diffusion-like equation:

$$\frac{\partial Q}{\partial t} - D\vec{\nabla}^2 Q = 0 \qquad (1)$$

where Q can be expressed as a function of time and position, i.e. $Q = Q(\mathbf{r}, t)$. Note that this equation has no homogeneous term because in our case the source is zero at $t = 0$ s. The coefficient D represents the propagation of the induced order. In order to satisfy our experiment at a temperature T, we choose a particular solution of equation (1):

$$Q(z, t) = \exp(-bt)[A\cos(Kz) + B\sin(Kz)] \qquad (2)$$

*Corresponding author. Email: pricardo@dfi.uem.br

Liquid Crystals
ISSN 0267-8292 print/ISSN 1366-5855 online © 2006 Taylor & Francis
http://www.tandf.co.uk/journals
DOI: 10.1080/02678290600583227

where A, B and b are constants that depend on the initial boundary conditions of the experiment, $K=(b/D)^{\frac{1}{2}}$.

From the boundary conditions of the experiment, one gets:

$Q(0, 0)=A=Q_0$, representing the maximum order;
$Q(L, 0)=0$, where L is the width of the container,
$B=-Q_0\cot(KL)$
$Q(0, t)=Q_0\exp(-\alpha t)\Rightarrow b=\alpha$.

The optical transmittance of the sample at a given temperature can be written as a function of $[Q(z, t)]^2$ [13] where in our case:

$$Q(z, t)=Q_0\exp(-bt)[\cos(Kz)-\cot(KL)\sin(Kz)] \quad (3)$$

A beam along the x-axis can experimentally produce this situation at a fixed frequency in the isotropic phase of a liquid crystal. The total optical transmittance from the sample can be expressed as [13]

$$I(t)\propto\int[Q(z, t)]^2\,dz \quad (4)$$

where the z-axis corresponds to the direction of the probe beam laser. The expression (4) has an adjustable parameter that can be obtained by fitting the experimental data. To solve equation (4) we used Mathematica 4.2 [14].

3. Experimental

The sample studied here was a mixture of potassium laurate (KL), decanol (DeOH) and water with concentrations, in percentage weight, of 26.89, 6.39 and 66.72, respectively. The phase sequence [15], determined by optical measurements, is Lamellar (L1) up to 19.0°C isotropic (I) between 19.0 and 34.0°C and Lamellar (L2) above 34.0°C. The lyotropic mixture is placed on a Hellma container (with dimensions $a=10$ mm width, $b=5$ mm thickness and $c=40$ mm height) between crossed polarizers. Figure 1 shows the experimental set-up. The laboratory frame is defined with $x//b$, $y//c$ and $z//a$. The sample is illuminated with a He-Ne ($\lambda=632.8$ nm) probe laser beam of 10 mW. The polarization direction of the probe laser beam is 45° to the y-axis. The external stimulus is produced by a pump laser beam (Nd- YVO_4, $\lambda=532$ nm) focused on the sample at normal incidence by a set of lenses. The power of the pump beam was varied from 0 to 2.0 W and the frequency of the pulse was controlled with a mechanical chopper. The pump beam is located perpendicularly with respect to the probe beam. The sample is placed on a controlled temperature system with 0.1°C stability. At room temperature, the transmittance of the lyotropic

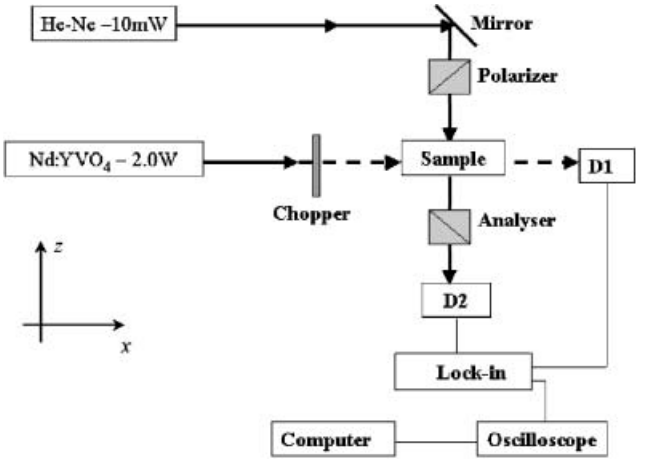

Figure 1. Sketch of the experimental set-up; D1 and D2 are photodiodes.

liquid crystal in the isotropic phase is detected with a photodiode connected to an oscilloscope. The reference signal is measured from the photodiode D1 (figure 1). The experimental values are stored in a PC by means of a GPIB interface. In order to obtain a better transmittance signal to noise ratio, a simple mean of three acquisitions was made. The same experimental procedure was applied to the whole range of temperature of the isotropic phase.

4. Results and Discussion

In order to verify the effect of induced order in the I phase of the lyotropic mixture by pulsed light, we used the experimental set-up described in § 3. The experimental procedure was as follows: The sample was placed in a controlled temperature system, with its temperature is maintained at the I phase. The sample was then illuminated with a polarizing laser beam; in this situation the measured optical transmittance, detected by the photodiode, corresponds to the transmittance arising from the disordered phases. When some order is induced in the I phase of the ternary mixture (KL/DeOH/water) an astonishing increase in the optical transmittance was observed. The external stimulus inducing order in the I phase was supplied by a pump laser beam of 2.0 W and frequency 130 mHz. For a pump beam with power lower than 1.5 W, no induced order was observed.

Figure 2 shows a typical optical transmittance of the sample as a function of time at 31.0°C (isotropic phase). When the pump beam (circle) is turned off the transmittance (triangle) of the sample decreases with a characteristic time of $\tau=18$ ms. The inset in figure 2 shows this decay process. This time has the order of

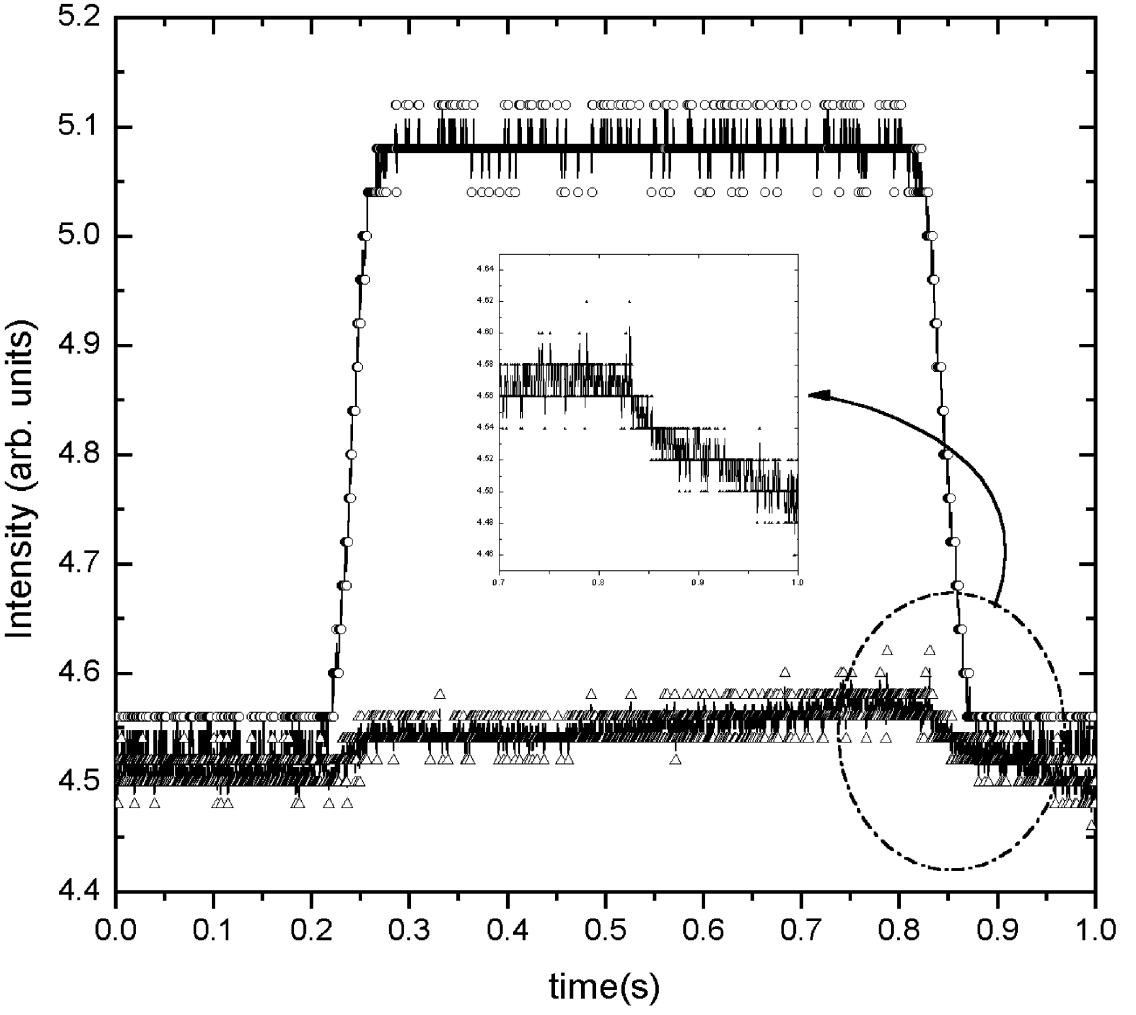

Figure 2. Evidence of order induced by pulsed light as a function of time in the isotropic phase of the lyotropic liquid crystal. $T=31.2^{\circ}C$; $P_{Pump}=2.0\,W$.

magnitude of the thermal effects. However, we emphasise that this effect is of a mechanical nature related to the pulsed laser beam; it does not appear when a continuous pump beam is used. We speculate that two phenomena can occur together when the sample is illuminated: the optical Kerr effect [16] and a thermal effect [17]. Our experiment does not separate these two effects. The induced order effect can also be interpreted as a transfer of momentum from photon to micelle [18].

We have experimentally verified that the induced order effect is optimized when the mixture is near to the nematic discotic phase (N_D). In order to amplify the effect shown in figure 2 we make a simple mean of five intensity measurements; figure 3 shows the amplified effect. The induced order in the disordered systems can be theoretically explained by equation (4). In this approach the order $Q(z, t)$ created by the pump laser beam is propagated into the isotropic phase. This order vanishes in space (z-direction) and decays exponentially as a function of time. The optical transmittance of the sample induced by light at a given temperature can be written as a function of $[Q(z, t)]^2$ integrated over the optical length (5.0 mm).

We note that equation (4) is very sensitive to the parameter b. According to our experimental results $b=1/\tau=55.5\,s^{-1}$. In this sense, the coefficient of the propagation of order, D, is the only adjustable parameter in equation (4) that agrees with the experimental data; $D=7.0013\times10^{-8}\,cm^2\,s^{-1}$. A variation of $3\times10^{-4}\,cm^2\,s^{-1}$ in D causes, at least, a deviation of 0.8% in equation (4), in comparison with the experimental data. From the order of magnitude of D we can conclude that the order induced inside a disordered phase by pulsed light is very fast (four orders of magnitude) in comparison with the order induced by mechanical stress, as obtained in [7].

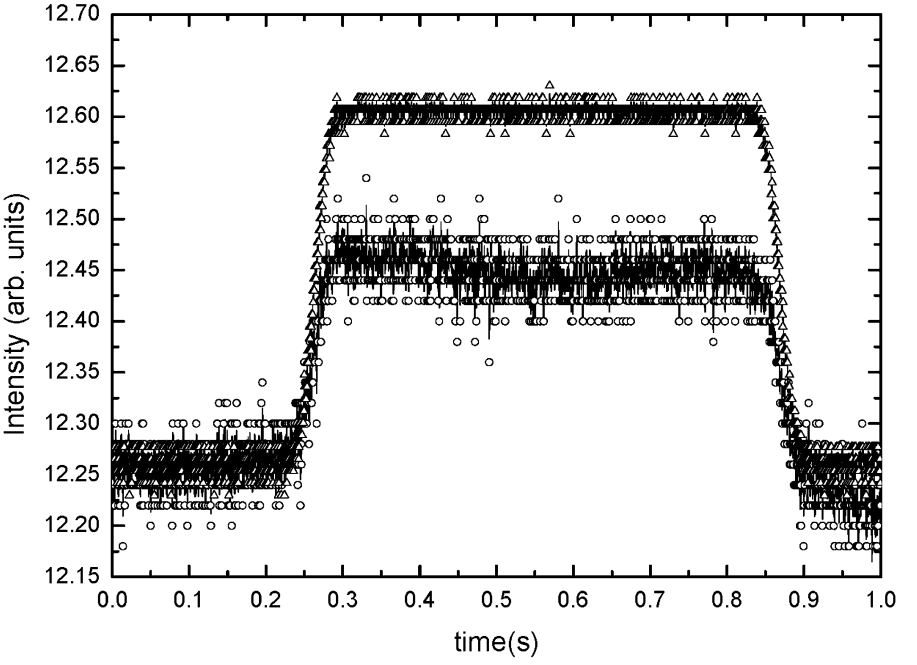

Figure 3. Amplified intensity as a function of time in the isotropic phase of the lyotropic liquid crystal. $T=31.2^{\circ}C$; $P_{Pump}=2.0\,W$. Triangles denote the reference signal, circles the optical transmittance signal.

5. Conclusions

In this work an effect of induced order by pulsed light, in the isotropic phase of a micellar lyotropic system is reported for the first time. This effect is explained in terms of a diffusion-like equation for the induced order parameter. From this approach the propagation of orientational order induced by light is of the order $10^{-8}\,cm^2\,s^{-1}$. This parameter indicates that the propagation of the induced order inside a disordered phase is faster by four orders of magnitude than the order induced by mechanical stress.

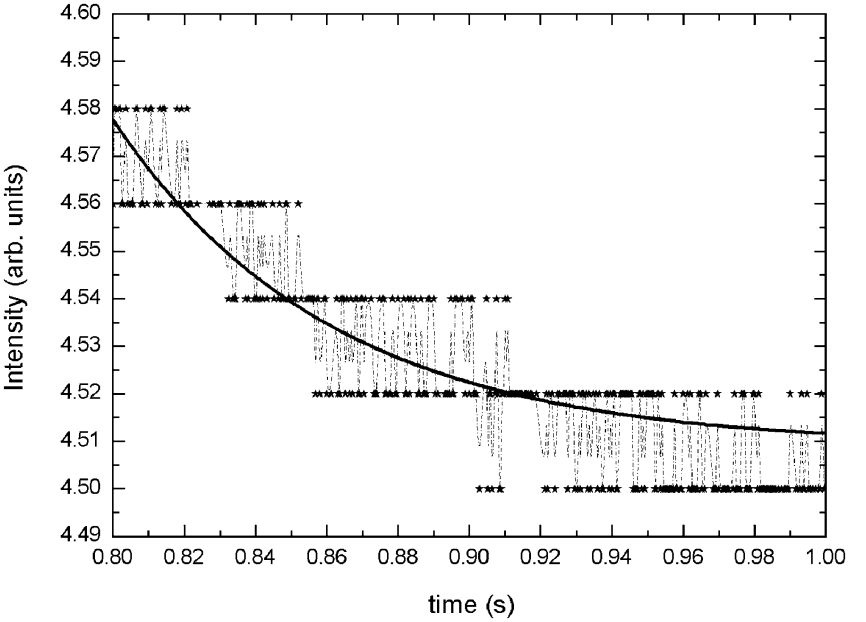

Figure 4. Theoretical fitting of the optical transmittance obtained from the propagation approach, equation (4); $D=7.0013\times10^{-8}\,cm^2\,s^{-1}$; $b=55.5\,s^{-1}$.

Acknowledgements

The authors acknowledge the Brazilian Agencies CNPq, FAPESP and Fundação Araucária for financial support. We also wish to thank Odair G. Martins for helpful discussions.

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