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ORIGIN OF OPTICAL SELF-PULSING AT A LIQUID CRYSTAL INTERFACE

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Abstract We discuss the dynamics of the light induced nematic-isotropic transition underlying the role that can be plaied by nuclei of critical size. It is found that critical time for droplet formation can have a very rare dependence on the overheating temperature which can explain the self-pulsing phenomenon recently observed at a glass-liquid crystal interface.

INTRODUCTION

Recently considerable attention has been devoted to the laser induced nematic-isotropic transition and several nonlinear optical effects such as bistability[1], self-transparency[2] and optical switching[3] have been observed using different geometries. Some models have been introduced to describe the growth of an isotropic droplet into a nematic phase and good agreement has been found with the experimental data under steady state conditions. A satisfactory description of the dynamics of this phenomenon appears to be more complicated to account for the observed transient effects. A detailed theory and experimental studies on the dynamics of the total internal reflection to transmission switching has been given for a glass-nematic interface without including the laser induced phase transition[4,5]. In the same geometry, near the nematic-isotropic transition an original self-pulsation effect has been reported[6]. The pulsation phenomenon has been explained with the formation of an unstable isotropic droplet for a particular impinging optical power.

In this paper we discuss the dynamics of the laser induced nematic-isotropic phase transition and point out that our model is able to give a semi-quantitative explanation of the observed self-pulsing effect. A comparison between the theoretical expectations and new experimental data on this effect are also presented.

The experimental geometry which we deal with is the one reported in ref. 6. The liquid crystal sample (the dyed nematic D101E63 by BDH) was placed between two glass prisms (n_p = 1.63), the surface of which had been treated to get a planar texture. The sample thickness was 75 μm, as fixed by Mylar spacers.

The incident light beam from an argon-ion laser (λ = 514 nm) was linearly polarized in the direction normal to the incidence plane in order to travel as a pure ordinary ray in the liquid crystal (n_o = 1.54) and focused by a lens (f = 100 mm) on the prism-liquid crystal interface. This configuration was chosen in order to avoid orientational effects and to have a positive nonlinear refractive index ($dn_o/dT > 0$). The doped sample was chosen to enhance the thermal effect by increasing the light absorption.

In this condition for an angle of incidence, at the prism-liquid crystal interface, $\alpha_o > \alpha_T = \sin^{-1}(n_o/n_p) \sim 71°$ the light was totally reflected.

An increase of the incident power, partially absorbed at the interface, produced a rise in the temperature sufficient to induce a phase transition to the isotropic state. As a result the critical angle α_T increased becoming larger than the fixed incident angle α_o, then switching from total internal reflection to transmission occurred.

When the laser beam was carefully focused onto the liquid crystal interface self-pulsing of the transmitted beam was observed in a narrow range of optical power near the switching power.

The features of the pulsing signal were the following: a) pulse duration was approximately 20 msec not dependent on the optical power; b) period of pulsation was very regular and dependent on the optical power in the range of 100-500 msec; c) at $\alpha_o \approx \alpha_T$ the period of pulsation was strongly dependent on the laser spot size on the sample (this new experimental result is reported in the last section).

Pulsation was observed only by increasing the optical power, i.e. in the switch-on process and it was not observed in the switch-off process.

DYNAMICS OF THE LASER INDUCED PHASE TRANSITION

We have already pointed out that light absorption in the nematic phase produces a temperature rise which may induce the nematic-isotropic transition in a portion of the liquid crystal material. The emergence of an isotropic phase will manifest itself in the creation of droplets (nuclei) of this phase, in their further growth and coalescence. As is well known[7], the kinetics of phase transitions is fairly complicated, therefore we shall not aim at describing all the above mentioned processes in detail. We will focus our attention to the determining role in the formation of the new phase played by the so-called nuclei of the critical size since all nuclei of smaller sizes are unstable and vanish. Actually the mechanism of formation of the critical-size nuclei can affect any transient phenomena occurring "near" the phase transition.

In the absence of any mechanisms diminishing the absorbed energy of the incident beam, the more the sample is being heated, the greater part of it will transit into the isotropic state. Thus, the laser beam will penetrate deeper and deeper into the sample, which in the final run will lead to a normal transmission through the isotropic nematic phase. Yet, if there is a mechanism owing to which the absorbed energy decreases due to the nucleation of the new phase, the system cools down. In virtue of this, the nuclei vanish and the system comes back to the original state.

It is evident that the characteristic times describing the dynamics of nucleation and the process of heat transfer play an important role. Let us perform the necessary estimation of these times.

To find the characteristic times of the heat transfer, let us make use of the thermal conductivity equation which for an isotropic medium reads:

$$\partial T / \partial t - \chi \Delta T = (\sigma / c_p)(|E|^2 cn / 8\pi) \qquad (1)$$

Here χ is the thermal diffusivity of the sample, σ the absorption coefficient, c_p the specific heat per unit volume, c the speed of light and n the refractive

index.

The right-hand side of this equation describes the heating of the medium by the laser beam: E being the optical field.

Unfortunately we do not know values of all physical quantities for the material, used in experiments[3,6], that is why hereafter we shall employ the characteristic values for regular nematics[8]. Since by order of magnitude these values are fairly close to each other, one can assume that we can use them for qualitative estimation.

From Eq. (1) one can get two characteristic times:

$\tau_L \sim L^2/\chi$ corresponding to the heating (cooling) of the sample thickness L

$\tau_d \sim d^2/\chi$ corresponding to the heating (cooling) of the beam-diameter area.

Inserting here the experimental values $L = 75\ \mu m$, $d \approx 35\ \mu m$, and $\chi \sim 10^{-3}\ cm^2/sec$, we shall get $\tau_L \sim 56$ msec, $\tau_d \sim 12$ msec. It becomes clear from these estimates that if nucleation occurred instantaneously, the time τ_L would characterize the transition from the nematic into the isotropic phase in the entire region of penetration of the laser beam through the sample. The time τ_d corresponds to the transition of this region into the nematic phase provided the energy pumping is absent (or decreases). In this case the cooling of the system down to the temperature of the medium, external with respect to the region of the beam, occurs due to the heat transfer in the direction orthogonal to the beam. The existence of finite characteristic times of the nucleation cardinally changes the situation.

The time of formation of a critical size nucleus, using formulas of ref. 7, is given by:

$$\tau_{cr} = A \exp (4\pi\alpha\, r_{CL}^2/3kT) \qquad (2)$$

where $r_{cr} = (2\alpha v/(v'-v)\delta T)\,(dp/dT)_o$ is the radius of the critical size nucleus.

Here $\delta T = T - T_o$, T_o is the phase transition temperature, T is the temperature of the metastable phase the nucleus is in equilibrium with, v' and v are the molecular volumes of nucleus and of the metastable phase, respectively, α is the surface tension on the boundary of the isotropic and nematic phases, k is the Boltzmann constant, p is the pressure.

The preexponential factor can be, by order of magnitude, represented by the expression A r^2_{cr}/D, where D is the diffusion coefficient.

Inserting into (2) the data for MBBA $\alpha = 2\ 10^{-2}$, erg/cm^2 (ref.9) $(v'-v)/v = 1{,}2\ 10^{-3}$ (ref.10), $(dp/dT)_o = 4\ 10^7$ dyne/cm^2 K (ref. 11), $= 5\ 10^{-7}$ cm^2/sec[6], we can get the dependence of τ_{cr} on the value of the overheating δT.

It is noteworthy that in virtue of the exponential character of Expr. (2) this dependence is rather rare. So, e.g., for $\delta T = 0{,}3$ K, τ_{cr} 6 sec, for $\delta T = 0{,}4$ K, $\tau_{cr} \sim 10^{-2}$ sec, for $\delta T = 0{,}5$ K, $\tau_{cr} \sim 5\ 10^{-4}$ sec.

Thus it is clear that at $\tau_{cr} > \tau_L$ there can be a situation when the sample already overheated by the depth L for the time τ_L the transition into the isotropic state is prolonged up to the times τ_{cr}.

Now estimate the temperature of the overheating of the sample depending on the power of the laser beam.

From eq. (1) follows

$$\delta T \sim (\sigma I/c_p \chi)(L^2/\pi^2)$$

here L is the characteristic length and $I = cn\,|E|^2/8\pi$ the light intensity.

Representing I in the form $I = 2P/\pi w_o^2$, where P is total input power, w_o diameter of the beam, we shall get

$$\delta T \sim (2\sigma P/\pi C_p \chi)(L/w_o)^2(1/\pi^2) \qquad (3)$$

Inserting here $c_p \sim 10^7$ erg/cm^2 K^4, $\sigma = 1$ cm^{-1}, L = 75μm we shall get δT 0,35K.

The above estimates for the time of formation of the critical size

nucleus at the overheating $\delta T \sim 0{,}35$ K yield $\tau_{cr} \sim 6{,}3\ 10^{-1}$ sec. which is in the range of the measured pulse period. It should be noted that the values of the physical parameters, used in the estimates for δT and τ_{cr}, do not correspond to the substance employed in experiments. Therefore the comparison of experimental data with the obtained τ_{cr} is not justified. Nevertheless, this study makes it possible to conclude that at certain pumping powers P the system in principle can be heated up to the temperatures at which the dependence τ_{cr} is very rare. Then small variations of δT can change the value of τ_{cr} by orders of magnitude.

Now finally formulate the proposed pulsation model. At the increase of the power of the incident beam the sample is heated up to the temperature exceeding the critical temperature, droplets (nuclei) of the nematic phase are created in the region of the size of the beam and as a consequence, the light penetrates through the sample. Yet at nucleation the scattering increases and thus the absorbed energy and the temperature of the medium diminishes. This change in temperature causes the collapse of the nuclei. The cooling of the entire region of the beam penetration due to the heat transfer processes occurs for the time $\tau \sim \tau_d$. At the cooling of the system down to temperatures $T \approx T_o$, the whole region transits into the nematic state and becomes no more transparent and the process of nucleation starts all over again. However, now it is our assumption that the temperature of the original state is close to T_o and it means that the effect occurs only for optical powers near the critical power for the TIR-transmission switching. In this way the determining role in this process is played by $\tau_{cr} > \tau_L$.

The proposed model also ensures the absence of pulsations when the beam power decreases. In fact in this case both the power decreases and the nucleation of the new phase (in this case - nematic) generates the fall of the temperature of the medium. Thanks to this, the formation of the low-temperature nematic phase is simplified.

COMPARISON WITH EXPERIMENTAL DATA

The comparison of the value of τ_d with the experimentally observed duration of pulses yields a fairly good agreement. For reasons given above

it is not possible to carry on a quantitative comparison between the value of τ_{cr} and the pulses period. Nevertheless it is possible to compare the expected dependence of τ_{cr} on the laser spot size and on the impinging optical power with the dependence obtained for the pulse period on these experimental parameters. In fact from the above mentioned expressions we have $\delta T \sim 1/w_o^2$, $\tau_{cr} \sim (\delta T)^{-1} \sim w_o^2$, thus:

$$\tau_{cr} = a w_o^4 \exp(b w_o^4) \tag{4}$$

where a and b are constant parameters.

Measurements of the period of pulsation has been performed for different distances between the focusing lens and the liquid crystal interface. The lens movement was controlled by a micrometer screw while the minimum spot size was measured and the other values were calculated according to the well known expression for a Gaussian beam.

Experimental data (dots) are reported in fig. 1 where $\ln \tau_{cr}$ is plotted vs w_o^4. Good agreement with the dependence given by eq. (4) is found (straight line in fig. 1).

A second test of the proposed model can be done by looking at the dependence of τ_{cr} on the pumping power.

In this case

$$\delta T \sim P \quad \text{and} \quad r_{cr} \sim P^{-1}$$

therefore

$$\tau_{cr} = a'/P^2 \exp(b'/P^2) \tag{5}$$

The period of pulsation have been also measured versus the laser power and data are reported in fig. 2 together with the expected behavior given by eq. (5). Fairly good agreement is found also in this case.

It is clear that to make more definite conclusions on the validity of the proposed model it is necessary to carry on additional experiments also using different liquid crystals where all the material parameters are known and performing measurement of δT in the region of propagation the beam. Nevertheless the comparison reported here between the available experimental data and the proposed model strongly support the idea of the determining role played by the nuclei of critical size.

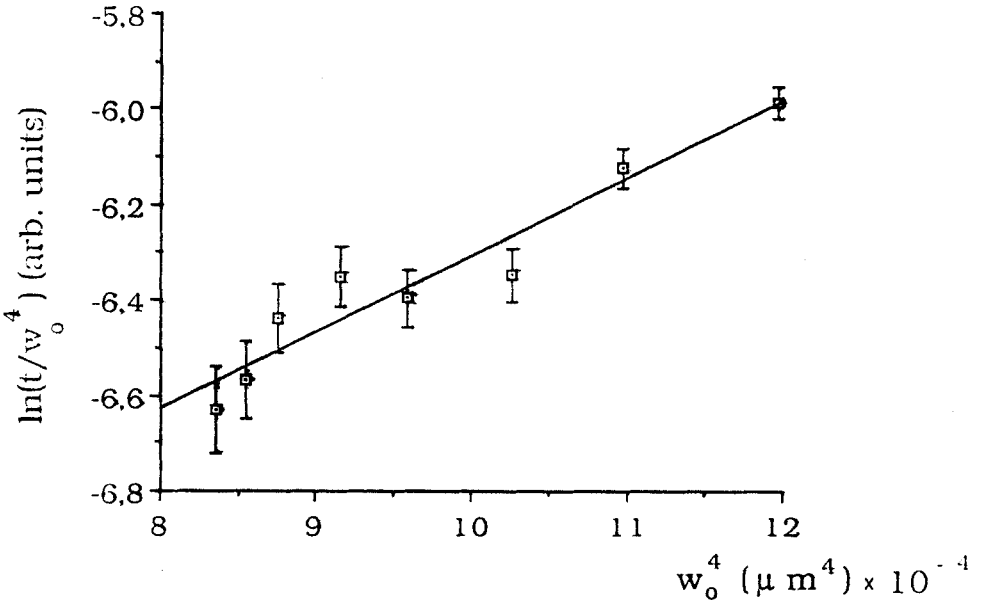

Fig. 1 - Dependence of pulsation period on the laser spot size. Continuous line is calculated from eq. (4)

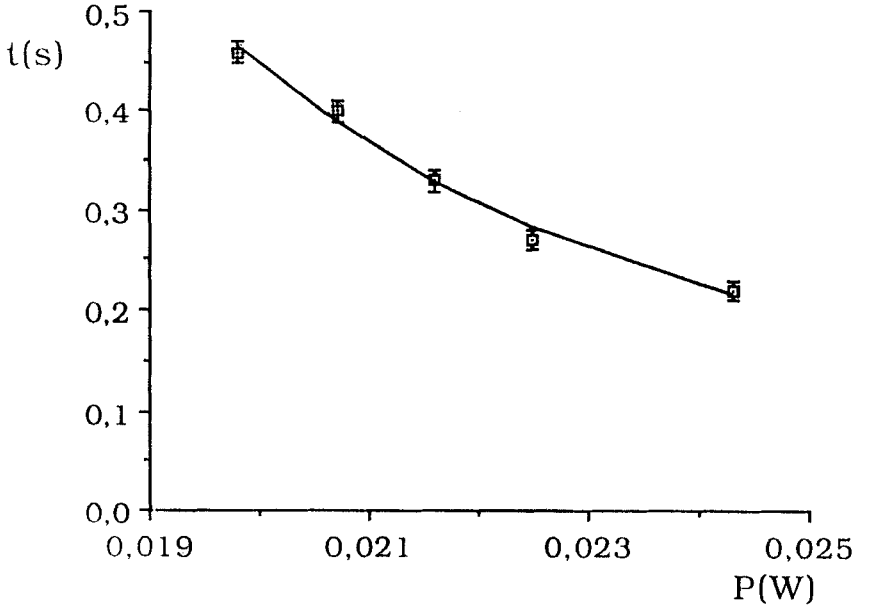

Fig. 2 - Dependence of pulsation period on the laser power. Continuous line is calculated from eq. (5).

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